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Topical review

Research frontiers in magnetic materials at soft X-ray synchrotron radiation facilities

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Abstract

Current and anticipated future research frontiers in magnetism and magnetic materials are discussed from a perspective of soft X-ray synchrotron utilization. Topics covered include dimensionality (including effects of spatial dimensions and differing time scales), magneto-electronics, structure/property relationships, and exploratory materials, with an emphasis on challenges that limit the understanding and advancement of these areas. Many soft X-ray spectroscopies can be used to study magnetism associated with transition and rare earth metals with element- and chemical-state specificity and large cross-sections associated with dipole transitions from $p \rightarrow d$ and $d \rightarrow f$ states. Established electron spectroscopies, including spin-resolved techniques, yield near-surface sensitivity in conjunction with linear and circular magnetic dichroism. Emerging photon-based scattering and Faraday and Kerr magneto-optical measurements can be used beyond the near-surface region and in applied magnetic fields. Microscopies based on either electron or photon spectroscopies to image the magnetization at 50 nm resolution are also emerging, as are time-resolved measurements that utilize the natural time structure of synchrotron sources. Examples of research using these techniques to impact our fundamental understanding of magnetism and magnetic materials are given, as are future opportunities. © 1999 Elsevier Science B.V. All rights reserved.

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1. Introduction

This report discusses current and anticipated research frontiers in magnetism and magnetic materials with an emphasis on the utilization of vacuum-ultraviolet (VUV) and soft X-ray

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techniques. The report grew out of a workshop in Berkeley, CA, in March of 1998 to explore new scientific directions with the unique capabilities provided by high-brightness, low-energy synchrotron-radiation facilities, such as the Advanced Light Source (ALS) at the Ernest O. Lawrence Berkeley National Laboratory. Select examples of past accomplishments within the field of magnetism are enumerated and emerging capabilities are identified. The report then addresses how the ALS can impact the frontiers of magnetism research and where the unique strengths and greatest opportunities of such facilities lie. Finally, it provides a road-map for future investment in research in magnetism and magnetic materials at ALS.

Interactions among electrons in solids give rise to many interesting physical properties that lead to such practical applications as superconductivity and magnetism. The consequences of magnetism and magnetic materials for society are enormous, ranging from recording heads and media in information-storage technology to components in the most basic transformers and motors involved in the generation and application of electric power. While magnetic materials have been used with increasing sophistication since the lodestone compasses of the Phoenicians, many basic questions about the microscopic physical interactions and properties in these materials remain unanswered. Increasingly sophisticated applications, driven in part by advances in the synthesis of complex structures, often with nanometer-scale dimensions, require increasingly sophisticated experimental techniques that can directly probe electronic and spin states as well as the magnitudes of atomic magnetic moments and the magnetic microstructures. Such basic properties are ultimately responsible for the remarkable usefulness of these materials.

In many ways, research in magnetism and magnetic materials is experiencing a renaissance [1] that has been enabled by developments in the last several decades in the fields of semiconductor physics and materials. The ability to control thin-film growth at the atomic level to form epitaxial and heteroepitaxial semiconductor structures has more recently been extended to magnetic nanostructures, including metallic, oxide, and semiconducting

phases. Fig. 1 shows a wide variety of atomically engineered magnetic nanostructures that can currently be grown and studied. This new era is driven both by the interesting physics of these materials, and by the \$50 billion per year magnetic-storage industry (\$150 billion per year if magnetic-recording tape, video, etc., are also included). This is in close analogy to the scientific impetus provided by the \$150 billion per year semiconductor industry. The pervasive role of hard and soft magnets in electric-power production and utilization also continues to motivate research at all levels into new materials that might save considerable amounts annually by reducing energy losses and saving natural resources consumed in the generation and use of electricity. The considerable relevance of magnetism and magnetic materials to society through both technology and economic factors cannot be disputed.

Many examples can be found to illustrate the close link between the fundamental physics of magnetic phenomena and their technological application; consider interlayer magnetic coupling in multilayer structures. Initial studies of coupling in multilayers were motivated primarily by scientific curiosity. (For an early claim of RKKY coupling in Cu/Ni see Ref. [2], for magnetic dipolar coupling in Mo/Ni superlattices see Ref. [3], for oscillatory coupling in Gd/Y superlattices see Ref. [4], for spiral coupling in Dy/Y superlattices see Ref. [5], for antiferromagnetic coupling in Co/Cu see Ref. [6] and for antiferromagnetic coupling in Fe/Cr see Refs. [7,8].) As giant magnetoresistance [9–11] (GMR) and the concomitant phenomenon of oscillatory interlayer magnetic coupling [12–14] were discovered, the technological relevance of these initially purely scientific discoveries [7,8] were brought to the marketplace as vital products within an extraordinarily short ten-year period. Fig. 2 shows a diagram of a magnetic recording head that uses a spin-valve read transducer based on the GMR effect. With the ability to synthesize novel magnetic films and nanostructures [15] exhibiting a variety of magnetic properties come prospects for new generations of devices. An example of an application based on magnetoelectronics [16], is the magnetic random-memory cells illustrated in Fig. 3; here, spin-dependent transport introduces

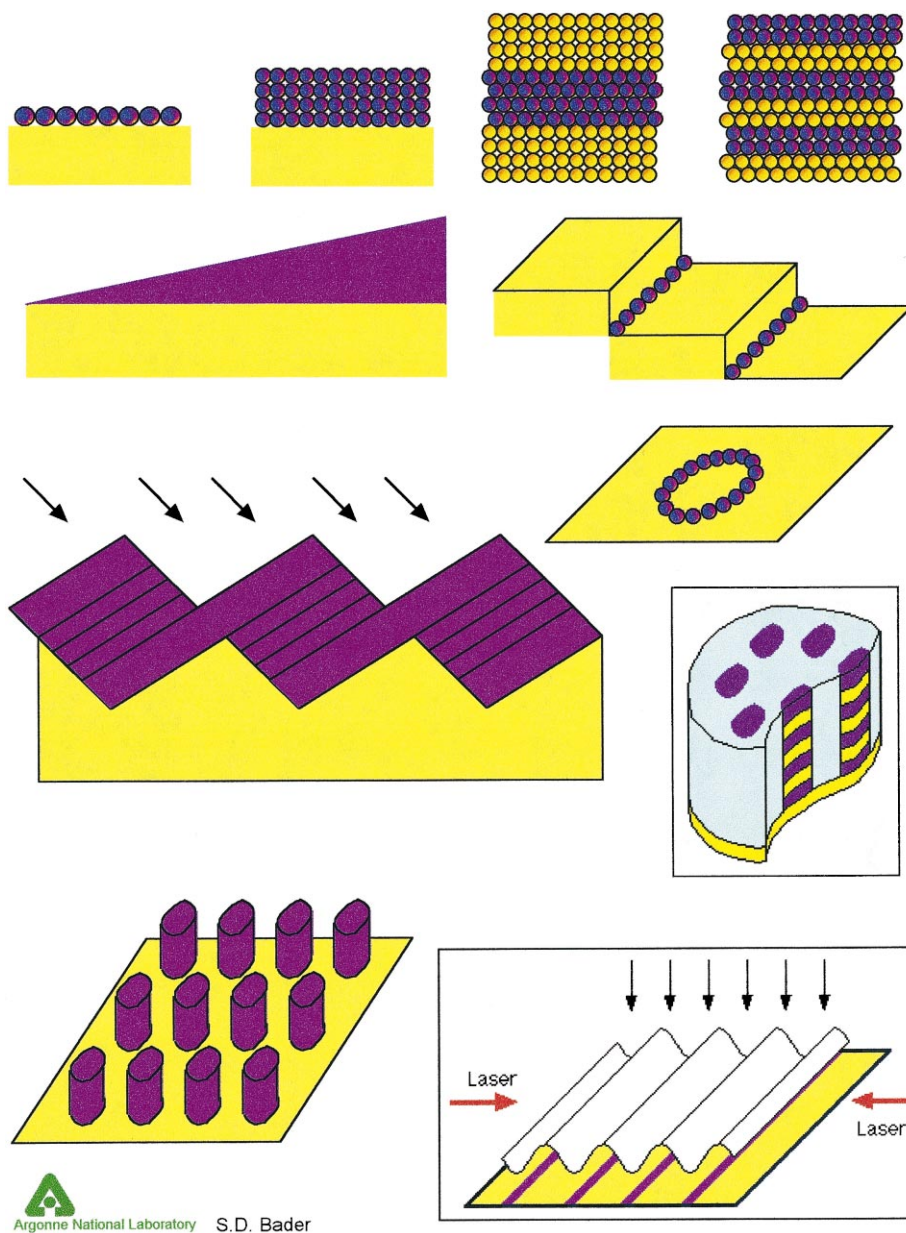


Fig. 1. Some geometrical arrangements of magnetic nanostructures of current interest are illustrated here. In general, dark features represent magnetic material. The top row indicates ordered monolayers and nanoscale thin films, sandwich structures, and multilayers. Wedged thin films present a range of thicknesses for study in a single sample. Decorated steps and quantum corrals have been grown with atomic-level control. Laterally patterned structures in the bottom include thin films, magnetic dots and wires, and arrays of magnetic-multilayer columns. The broad range of materials that can be grown in these nanostructures present many interesting fundamental questions and potential technological applications. They also present many experimental challenges, such as distinguishing magnetism in buried ultrathin layers from that at the interfaces between them (figure courtesy of S.D. Bader, Argonne National Laboratory).

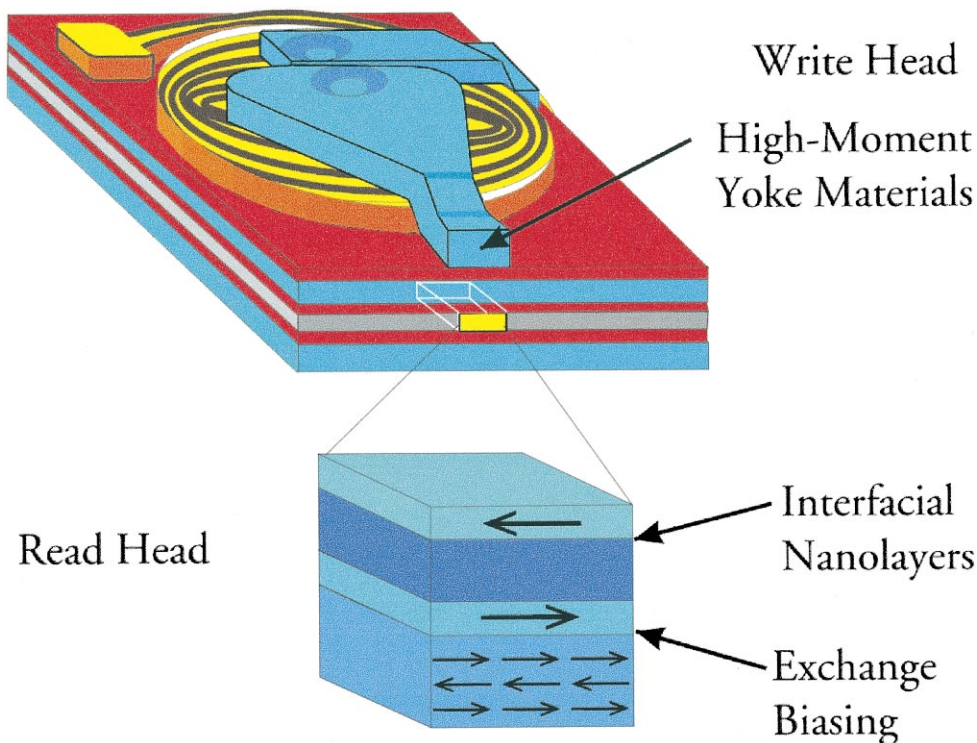


Fig. 2. Magnetic recording heads typically consist of a write head and a read head within the same lithographically defined structure. The write head consists of a coil and a yoke that guides the magnetic flux created by the coil to a pole tip. The large magnetic field emerging from the pole tip is used to write the magnetic bits into a thin magnetic film on a rotating magnetic-recording disk. The read head is used to retrieve the information written on the disk. It senses the magnetic flux emerging from the transition regions between the bits on the disk (see Fig. 12). In the spin-valve giant-magnetoresistance (GMR) head shown here, the flux from the disk is large enough to change the magnetization direction in one of the ferromagnetic layers which comprise the head. The magnetization direction in the second ferromagnetic layer is pinned by exchange coupling to an antiferromagnet and does not rotate. Owing to the so-called GMR effect, a sense current flowing through the spin valve experiences a resistance that is higher by about 10% when the two ferromagnetic layers are magnetically aligned antiparallel rather than parallel (figure courtesy of J. Stöhr, IBM Almaden Research Center).

magnetism into the realm of electronic devices, with the promise of large-scale nonvolatile memory. However, there is still no accepted detailed theoretical model for the mechanism of this effect.

In analogy with semiconductor materials, modern magnetic materials have complex structures with important functional properties over multiple length scales, ranging from the atomic to the macroscopic level. (See, for example, articles in Ref. [17] and see, for example articles in special issue in Ref. [18].) Moreover, experimental and theoretical efforts have shown that these properties evolve within time scales spanning femtoseconds to microseconds to hours and beyond. For example, not

only are magnetic phase transitions and moments (both spin and orbital) important parameters, but their dynamical behavior at nanometer length and ultrashort time scales can determine macroscopic properties. Yet the underlying physics in many cases remains largely unexplained. Structural complexity can take many forms. Nominally homogeneous binary alloys of magnetic 3d transition elements can exhibit Invar-like phase transitions linking structure and magnetism as electron–electron interactions change with composition see Ref. [19] and related articles in the same issue. Many important magnetic materials contain three or four different elements in structures with quite large unit

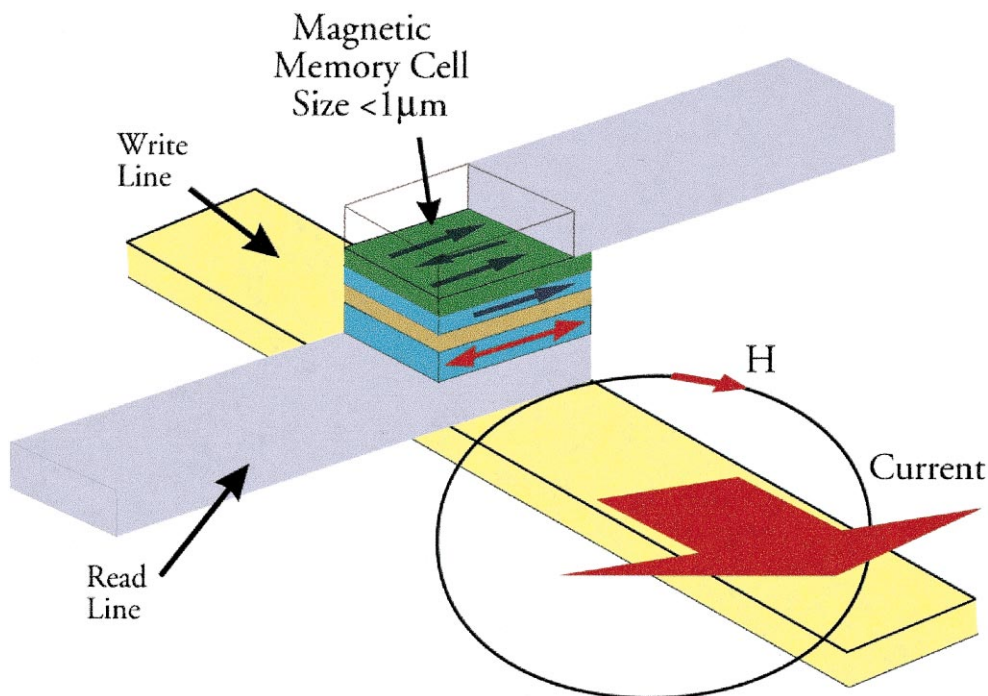


Fig. 3. Schematic illustration of a magnetic memory cell. The cell consists of a tunnel junction in which two ferromagnetic layers (e.g., cobalt) are separated by an insulator (e.g., Al_2O_3). The tunnel current flowing through the read line senses a resistance that depends on the relative orientation of the two magnetic layers, i.e., whether they are parallel (1) or antiparallel (0). As in the spin valve shown in Fig. 2, the magnetization direction in one of the magnetic layers is pinned by exchange coupling to an antiferromagnet. The magnetization direction in the other magnetic layer can be rotated by the magnetic field of a current flowing in a nearby write line (figure courtesy of J. Stöhr IBM Almaden Research Center).

cells, such as NdFeB hard magnets [20] and the manganites that exhibit colossal magnetoresistance (CMR). (For a review, see Ref. [21] and for a review of metal–insulator transitions in manganites, see Ref. [22].) As in the related cuprate high- T_C materials, interesting properties in these complex structures appear related to anisotropies and inhomogeneities inherent in the atomic arrangements within their unit cells. Increasingly common are heterogeneous structures consisting of finely layered or granular films of similar or different phases that themselves may be simple or complex. In these heterogeneous materials, anisotropy associated with size or structural aspects of the interfaces are likely to control the magnetic properties of interest. For an interesting recent example of enhanced magnetoresistance in insulating granular systems, see Ref. [23].

In general, the complexities faced in magnetic-materials research are frequently due to coupling of structural and electronic degrees of freedom over many scales. This results in intimate relationships between the structural, magnetic, and optical properties, which are increasingly utilized for new technologies — generating applications in sectors from communication to sensors to electro-optics to magnetic recording. The consequences of couplings between spin, charge, and lattice degrees of freedom are apparent in both inorganic and organic materials. Indeed, even opportunities for hybrid organic–inorganic materials and multifunctional designs are now emerging in the literature. Many of these strategies are evident in biological materials and are inspiring exciting new directions in materials design and synthesis of soft and biomimetic materials, starting from the molecular level. The

emergence of such materials provides new opportunities for studies that shed light on the fundamental mechanisms governing spin-polarized transport in low-dimensional systems, carrier-mediated magnetism, and magnetic-field-induced carrier localization. Such spin-engineered materials serve as model systems to test fundamental questions in quantum transport theory and local magnetism. For example, recent attempts to bridge the fields of semiconductor physics and magnetism have developed the basis for a new class of ‘spin electronics’ with fascinating technological opportunities [24]. Moreover, a number of theoreticians are posing scenarios in solids where spin and charge dynamics may be decoupled, with resulting predictions of new mechanisms for intrinsic spin transport.

The diversity of interactions underlying magnetic phenomena require probes that are sensitive to the local atomic and extended electronic structure (including electron spin), as well as to the geometric structure, in varied materials and over a wide range of spatial and temporal dimensions. Both lateral and vertical (depth) dimensions must be probed. Many experimental probes and techniques, such as magneto-optics with lasers [25], magnetometry, Mössbauer, electron spin resonance, neutron scattering, electron microscopy, and computational modeling, can currently provide information bearing on the questions of interest [17]. In roughly the last decade, techniques based on synchrotron radiation, such as spin-resolved photoemission, X-ray magnetic dichroism, and magnetic X-ray scattering, have been shown to provide unique capabilities for the study of magnetic phenomena and magnetic materials. Spin-resolved valence-band photoemission has provided unique insight into the mechanism of oscillatory exchange coupling in magnetic multilayers [26,27] and proven the existence of half-metallic ferromagnets [28] where only one spin subband contributes to electron transport. X-ray magnetic circular dichroism (XMCD) spectroscopy in the hard [29] and soft [30,31] X-ray range, through quantitative sum-rule separation of spin and orbital moments [32–35], has given us a clearer picture of the origin of the magnetocrystalline anisotropy [34,36] and revealed the existence and size of interfacial magnetic moments in

‘non-ferromagnetic’ metals, such as platinum [37] or copper [38,39], when adjacent to a ferromagnet. Spin-resolved core-level photoemission [40–44] has provided an element-specific probe of short-range magnetic order and magnetic phase transitions near surfaces. Hard X-ray magnetic scattering techniques are being extended into the soft X-ray to probe magnetic structure at nanometer lengths and above [45–48]. Faraday and Kerr magnetooptical rotation techniques common in near-visible regimes are likewise being extended into the soft X-ray, gaining element specificity and resonant signals much larger than at longer wavelengths [49,50]. Emerging techniques for soft X-ray magnetic microscopy [51] utilize several of these effects for elemental, magnetic contrast. Together these capabilities have clear potential to impact the study of magnetic materials and nanostructures in many ways.

2. Research frontiers in magnetic materials and phenomena

While it can be risky to try to identify long-term future trends in any scientific discipline, the process of doing so sensitizes the community and challenges us to transcend the known and the obvious, so as to go beyond the mainstay incremental advances and suggest potential new paradigms of the future.

The broad scientific issues that will form the fabric of the future can be categorized in many ways. Several defining attributes of magnetic materials cut across any classification scheme and are central in current and likely future research. Examples include the spin-polarized electronic structure, magnetic anisotropy, and the phase transitions that result in ferromagnets, antiferromagnets, and ferrimagnets. Underlying these attributes is the paradigm by which magnetic interactions and phenomena can be understood in terms of the geometrical and electronic structure that are inseparably linked in host materials. Table 1 categorizes research trends in magnetism emphasizing: (1) the underlying role of dimensionality [1,52], including both spatial and temporal degrees of freedom, (2) the emergence of magnetoelectronics [16],

Table 1

Current and future trends in magnetism and magnetic-materials research

Dimensionality—space and time

Magnetic anisotropy
Domain walls, domain correlations
Magnetic dynamics/fluctuations/melting
Weakly interacting systems
Phase transitions
Quantum tunneling

Magnetoelectronics

Spin injection and transport
Quantum confinement
Magnetic semiconductors
Disorder

Structure and magnetic order

Magnetic anisotropy
Frustration
Proximity effects
Disorder
Interfacial effects

Exploratory materials

Hybrid structures/competing interactions/frustration
Active interfaces
Biomagnets
Molecular magnets

including spin injection and transport in heterostructures that can serve in nonvolatile memory and logic-element arrays of the future, and magnetic semiconductors that may integrate spin-dependent transport and magneto-optics into semiconductor devices [53], (3) the broad fundamental materials-science area involving correlation of structure with magnetic properties [52,54–57], and (4) the exciting area of exploratory materials. Within each broad category are examples of areas perceived to provide opportunities for new fundamental understanding that may impact the science and technology of magnetism. In many cases, the same or related topics appear under different categories, indicating in part the pervasive role of the essential features of magnetic materials mentioned above.

2.1. Dimensionality — space and time

In magnetic materials, the spatial dimensions of interest range from the intra- and inter-atomic

scale, defining how electrons within and between atoms interact, to the macroscopic size of the sample as controlled by magnetostatic interactions. Research challenges range over this entire scale, from understanding how spin and orbital moments contribute to properties in specific materials to micromagnetic modeling of the magnetostatic interactions so as to yield both equilibrium properties and switching rates. Time scales of interest range from femtoseconds (and shorter) relevant to electronic transitions and electron–electron scattering to the very long times over which materials retain properties of interest. Switching of domains occurs by processes at intermediate time scales in the nano- and picosecond range. The ability to control spatial dimensions of magnetic features at the nanometer level, as illustrated in Fig. 1, opens the study of the dependence of all fundamental magnetic interactions on this important new experimental variable. Spatial and temporal dimensions are necessarily coupled, and as dimensions are reduced, the relative importance of different mechanisms for controlling dynamics changes. For example, of key importance in future magnetic-storage technologies is the ‘superparamagnetic limit’ at which the inherent magnetic anisotropy of a small magnetic particle is no longer strong enough compared to thermal energies to yield stable magnetization over the extended times needed in nonvolatile magnetic memory [58]. The prevalence of buried, ultrathin layers and interfaces in magnetic nanostructures or nanoparticles underscores the fact that the controlled growth and characterization of buried interfaces remains one of the great experimental challenges in materials science, as discussed in Section 2.3 below. In many cases, these magnetic nanostructures represent model systems for the study of interesting physical phenomena, while at the same time having direct relevance to applications.

2.1.1. Anisotropy

Magnetic anisotropy ultimately derives from the symmetries defining magnetic interactions, and remains at the heart of current research both from fundamental and applied materials perspectives. Fig. 4 shows magnetic anisotropy constants for a variety of materials and reveals this link between

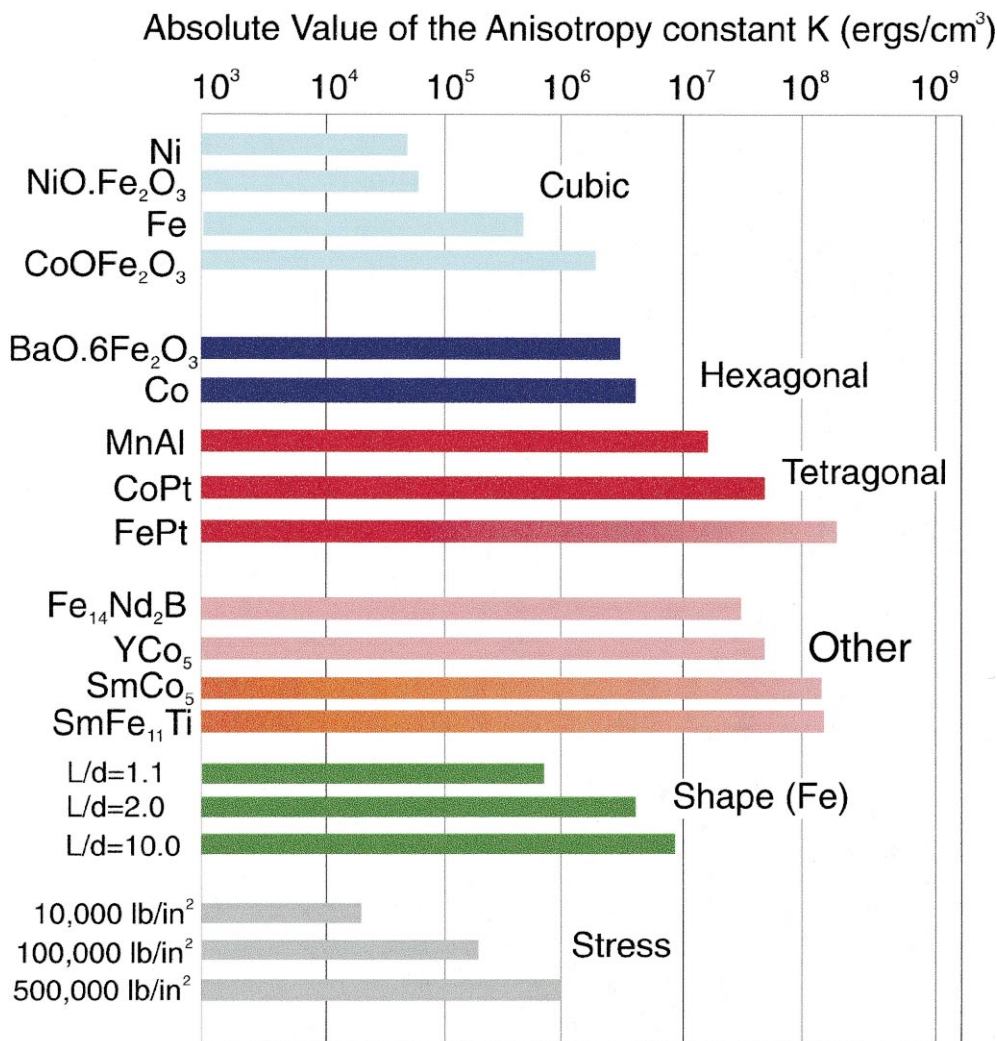


Fig. 4. Anisotropy constants for various magnetic materials show trends that reveal correlations between atomic structure, crystal structure and magnetism. Anisotropy is a fundamental property defining the suitability of magnetic materials for different applications whose microscopic origin is still being unraveled. Experimental probes that are sensitive to the spin-resolved electronic states underlying anisotropy in complex materials are needed to explain these trends at a microscopic level (figure courtesy of D. Weller, IBM, based on data taken from B.D. Cullity, *Introduction to Magnetic Materials*, Addison-Westley, Reading, MA, 1972, p. 381, and T. Klemmer et al., *Scripta Metallurgica et Materialia* 33 (1995) 1793).

anisotropy energy and symmetry through crystal structure. Anisotropy is known to have two major, often competing contributions. One is the dipolar coupling between individual atomic spins, which in practice leads to a dependence of the anisotropy on the macroscopic shape of the material (shape anisotropy). The other is the coupling of the electronic

spin to the lattice via the spin–orbit coupling (magnetocrystalline anisotropy). Because of the difficulty in separating various anisotropy contributions and the small size of typical anisotropy energies (10^{-4} – 10^{-6} eV/atom), the microscopic origins of anisotropy in specific materials generally remain poorly understood. The in-plane anisotropy typically

exhibited by thin magnetic films has found great utility in magnetic recording, yet films exhibiting perpendicular magnetic anisotropy are highly desirable for some applications, such as magneto-optical recording (see, for example Ref. [59]). Some thin films (e.g., a-TbFe, CoPt, and FePt) are found or can be grown to exhibit perpendicular magnetic anisotropy [57], and some nanoscale epitaxial films (e.g., iron films) exhibit reorientation transitions from in-plane to out-of-plane magnetization with changing growth conditions [60–64].

The ability to control the dimensionality of magnetic structures opens up the field of tuning magnetic properties by engineering anisotropy. Below are three examples based on effects resulting from layered or multilayered films in which the thickness of individual layers is of order 1–10 nm [65].

- Nonmagnetic materials (e.g., gold, platinum, and palladium) interleaved between soft ferromagnets (e.g., cobalt) yield new composites with unique magneto-optic properties, namely perpendicular magnetic anisotropy [66,67]. Understanding the origin of the magnetic anisotropy and magneto-optic response requires a rather detailed understanding of the interfacial region in terms of possible intermixing and topological roughness, in addition to the inherent asymmetry imposed by the interface.
- When a ferromagnet is grown in a magnetic field on top of an antiferromagnet, the magnetization direction in the ferromagnet becomes unidirectionally pinned [68] (for a recent review see Ref. [69]). This phenomenon of anisotropic exchange biasing is not understood, despite the fact that it is already utilized in the manufacturing of magnetic recording heads. Understanding the mechanisms of exchange biasing again requires the detailed magnetic and structural characterization of buried layers and interfaces between the two materials.
- New permanent-magnet nanostructures can potentially be tailored with maximum energy products exceeding that of NdFeB by factors of two or more [70,71]. These exchange spring magnets comprise hard, permanent-magnet layers contributing high anisotropy and coercivity, owing to the presence of a rare-earth component, and soft

layers, which could be as simple as elemental iron, contributing a high magnetization per unit volume. The resultant coupling ideally yields high magnetization and coercivity. After magnetization, a reversed applied field influences the soft component first, but turning off such a field allows the system to spring back to the original state of full remanence, hence the term ‘spring’ magnets to denote these remarkable, newly introduced structures. The physics of the coupling across coherent interfaces, the spin dynamics, and switching processes offer new challenges to and rewards for understanding the energetics of magnetic-domain walls in a fundamental way. The layered nanostructures provide model systems [72] to test concepts that may open major vistas for the miniaturization of electrical motors and the benefit of energy efficiency both in electricity and fossil-fuel utilization. The latter is due to the preponderance of electrical motors in automotive and other transportation vehicles.

Similar strategies of combining high-anisotropy, hard magnets with soft magnetic layers are relevant in the quest to overcome the superparamagnetic limit in conventional magnetic-recording media, as discussed in more detail in Section 2.1.3 below. Thus, the concept of engineering anisotropy through judicious choice of magnetic nanostructures is creating a paradigm shift in our thinking about diverse magnetic applications that affect society in fundamental ways.

2.1.2. Domain walls/domain correlations

Domain walls themselves are systems of reduced dimensionality that are rich in physics yet exceedingly difficult to characterize on a microscopic scale. As layer thickness and lateral dimensions of nanostructures decrease below the length scales associated with domain walls, dimensional effects alter domain and domain-wall configurations. Understanding the changing energetics, structure, pinning, and dynamics of domain walls in systems with reduced dimensionality is important, and is especially challenging in buried layers of nanostructures. In particular, there are a number of outstanding issues concerning the spatio-temporal profile of a moving domain wall in real materials,

where nonlinear effects become important near the Walker limit, the magnetostatic mode associated with the shape of the magnetic domain and the speed of sound. While classical mechanisms, such as wall–wall interactions, need to be understood and controlled for technology, a number of fundamental physics questions on the macroscopic quantum tunneling of domain walls and correlation measurements need to be addressed, with their answers possibly providing solutions to existing limits of high-density information storage.

The spatial correlation of magnetic domains within and between the magnetic layers of layered structures is of fundamental importance for many applications of spin-conductance devices, whether for magnetoresistive, spin-tunneling, or spin-transistor operation. For systems where magnetic-domain sizes are large compared to the relevant spin-flip path length, it is the orientation of the local relative magnetic moment that defines the local and global conductance behavior [73]. If domain sizes are comparable to or smaller than this path length, more complex behavior is expected. For a meaningful comparison between the measured and calculated spin conductance, a quantitative description of the applied-magnetic-field dependence of these magnetic-domain correlations is required. Experimental measurements of domain correlations within these layered structures are a great challenge and generally require development of new measurement techniques with spatial and chemical sensitivity at the appropriate length scales. Advances in micromagnetic modeling of these complex structures also require reliable experimental data.

2.1.3. *Magnetic dynamics/fluctuations/melting*

Magnetic dynamics includes both magnetization-reversal processes (whether dynamic or adiabatic) and important issues associated with noise inherent in magnetic devices. Fluctuations can be associated with the reversal process and also with magnetic phase transitions. Since all magnetic order results from temperature-dependent phase transitions, the term ‘melting’ applies very broadly to the loss of order with temperature. Another important distinction is that between long-range magnetic order (e.g., as manifested in macroscopic

susceptibility) and short-range magnetic order (as now thought to be important in such phenomena as high-temperature superconductivity and CMR). All of these fundamental dynamic properties in turn depend on the dimensionality of systems of interest, and so they present continued opportunities for fruitful research in the context of nanostructures. For example, in a system with nanometer dimensions, there is a merging of long- and short-range order. Some specific questions associated with the picosecond time scales for such processes are considered below.

Traditionally, magnetization processes have been studied only at the nanosecond time scale or longer. Studies at shorter time scales have been precluded by induction, which limits both the speed with which one can increase the magnetic field and the speed with which one can measure the change in magnetization by an induced current in a pick-up coil. With the advent of powerful pulsed light sources, such as lasers and synchrotrons, magnetization can now be measured either by magneto-optic effects or by observing the spin polarization of electrons ejected from the magnetic material on a time scale that is limited only by the length of the light pulse.

The femtosecond time scale is important in the study of electron–electron scattering [74] and is presently reserved for fast pulsed lasers [75,76]. The picosecond time scale, however, should be accessible with the pulsed light emitted from synchrotrons, and there are important reasons to study magnetism on this time scale. One is to observe directly the postulated spin blocks formed at the magnetic transition in two-dimensional objects, such as thin films. Such spin blocks are spontaneously magnetized regions with a diameter of microns or less that form and dissolve on a time scale of picoseconds. It has not yet been possible to observe such spin blocks directly, but indications of their presence appear mostly in neutron-scattering experiments, where they are seen as enhanced scattering (critical scattering). Picosecond magnetization measurements (if possible with element sensitivity) would make it possible to directly determine the magnitude of the spontaneous magnetization in the spin blocks. More generally, picosecond studies of the motion of the

magnetization vector when external conditions, such as applied magnetic field, temperature, or pressure are changed, are of interest.

Of particular interest is the dynamics of magnetization reversal [77], because it is the process by which magnetic bits are written into recording media. The industry needs to reduce the time for writing a bit from its present value of 10 to 1 ns. Experimental studies of magnetization reversal are generally not in agreement with the familiar model of coherent spin rotation. Instead, complex processes, such as curling and buckling, are observed [78]. However, if the magnetic-field excitation occurs at an angle to the magnetization direction and on much shorter time scales in the picosecond range, such complexity is suppressed and reversal should simply follow the model of coherent spin rotation. The time for the elementary process of magnetization reversal is given by the Larmor precession in the anisotropy field and lies in the picosecond regime. Therefore, field pulses of a few picoseconds duration with peak amplitudes up to 2 T are required to study magnetization reversal.

So far, such field pulses can only be produced by electron bunches of high current density passing through the sample. The remanent magnetization pattern generated in a premagnetized sample by the field pulse can be imaged by magnetic-microscopy techniques. The results show that the application of field pulses as short as a few picoseconds produce coherent rotation of the magnetization. The process of magnetic recording can thus be made faster by several orders of magnitude [79]. The comparison of the observed magnetization pattern with a micromagnetic model yields information on the damping behavior after application of the field pulse. The study of the damping and its understanding on a quantum-mechanical level is a presently unexplored field of crucial importance for increasing the data rates in magnetic recording.

2.1.4. Weakly interacting systems

As with all electronic interactions, the correlations among spin-polarized states depend on dimensionality. Low-dimensional systems containing one- and two-dimensional magnetic states exhibit exciting properties of both theoretical and potentially practical interest. Especially interesting are

states consisting of half-integer spins. Long ago, Bethe demonstrated that the spin- $\frac{1}{2}$ Heisenberg chain would not order because of the presence of quantum fluctuations [80]. More recently, Haldane has conjectured that half-integer spin chains possess an excitation spectrum with an energy gap [81], whereas integer spin chains are gapless. Similar fascinating properties are shown by two-dimensional systems, such as the $S = \frac{1}{2}$ square copper-oxygen layers that are present in high- T_c superconductors. An intermediate case is represented by spin ladders, which are arrays of coupled chains [82]. More complex systems involve quantum chains and quantum ladders. The static and dynamical properties of such systems are of fundamental interest because they may relate to the occurrence of superconductivity and colossal magnetoresistance.

Fundamental physics and potentially significant practical applications are joined in the naturally layered magnetic structures, represented by the colossal magnetoresistive (CMR) manganites of the Ruddlesden–Popper phases $\text{SrO}(\text{LaMnO}_3)_n$, where SrO represents a blocking layer that separates n layers of perovskite-based octahedra [83,84]. The trivalent lanthanum sites are partially substitutionally filled by a divalent species, such as strontium, to realize a mixed-valent state in manganese that is characteristic of double-exchange ferromagnets. Even though double exchange is an almost 50-year-old concept [85], the resurgence of interest in these materials is due to their being candidates for magnetoresistance materials, being half-metallic ferromagnets with possible 100% spin-polarized carriers, and to their forming compatible junctions with high- T_c cuprate superconductors [86]. These phases are analogous to those encountered in the high- T_c cuprates, as well as in the exotic ruthenate materials [87]. Research opportunities include the manipulation of dimensionality and structural anisotropy to (1) tailor the magnetotransport properties and (2) to explore the underlying physics of double-exchange, polarons, short-range magnetic order above the Curie temperature, the competition with antiferromagnetic superexchange interactions, and the charge- and orbital-ordering states that occur at special compositions. Characterization of these complex

materials demands techniques that can identify local structure (and occupancy) [88], valence, magnetic moment, and magnetic anisotropy for each constituent with element and site specificity. In short, what is needed is a complete atomic, electronic, and magnetic structure description that includes each constituent in a structure which may contain 10–100 atoms per unit cell. While this may be a daunting goal, many new spectroscopic and structural techniques will be employed to elucidate the physics of these materials. It is a challenge to the collective imagination of the research community to reach the goal of fully understanding these wonderfully complex and potentially very useful materials.

2.1.5. *Phase transitions*

As with all phase transitions, magnetic phase transitions have a pronounced dependence on dimensionality. Magnetic nanostructures containing more than one distinct magnetic component bring both dimensionality and competing interactions together in the study of magnetic phase transitions. Distinguishing between the responses of different constituents in these phase transitions presents experimental challenges. It is likely that pulsed synchrotron radiation may be used to drive phase transitions in magnetic materials as a means of probing magnetic behavior and local interactions with unprecedented resolution. This class of nonequilibrium dynamics is largely unexplored, and the ability to induce transitions in magnetic moment with chemical specificity offers a unique opportunity to examine this behavior in low-dimensional materials. The possible role of quantum fluctuations and tunneling in phase transitions is an emerging area that appears rich in future opportunity.

2.1.6. *Quantum tunneling*

The process of miniaturizing magnetic materials has unexpectedly revealed fascinating new intrinsic classical and quantum-mechanical phenomena. Even the simplest magnetic system, the isolated single-domain particle, exhibits a wealth of exotic behavior that pushes us to the limits of our present understanding of the fundamentals of magnetism. The simple picture of classical magnetism suggests that the magnetic orientation of a small particle will

remain stable in one of two orientations indefinitely below the blocking temperature, defined as the temperature at which thermal fluctuations are energetic enough to overcome the volume anisotropy energy. However, quantum mechanics tells us that the states in the two potential wells can be coupled by tunneling, thereby leading to strikingly different dynamics. Several recent experiments have demonstrated the presence of these quantum dynamics in small systems and raised tantalizing questions ranging from the fundamental limits of information storage to the observation of macroscopic quantum phenomena [89]. One-dimensional barrier penetration is a misleadingly oversimplified depiction of quantum tunneling of magnetization — there is no simple Schrödinger equation that describes this process, since it is not an elementary particle that is tunneling, but a collective coordinate, the magnetization direction of a collection of spins. The area of magnetic quantum tunneling offers exciting new theoretical and experimental opportunities for research including the role of dissipation in quantum magnetic phenomena and quantum-measurement theory.

It might appear that this process points to limitations and restrictions on the use of small magnetic particles for the storage of information, since the switching of magnetic domains depends on a myriad of detailed features of the particles, and quantum effects ultimately limit the length of time that a magnetic bit can remain stable (e.g., through the superparamagnetic limit). Nevertheless, it seems equally plausible that these investigations will provide fundamentally new ways of using magnetic structures in technology. For example, theoretical investigations of magnetic quantum tunneling led to the surprising discovery that a selection rule quite generally forbids quantum tunneling for particles with an odd number of electrons! [90]. Thus, limitations imposed by quantum mechanics may be overcome. Another area for new research concerns the use of magnetic systems not for memory, but for logic. Some computational problems can be greatly accelerated in a ‘quantum computer’ in which bits and gates are implemented at the level of individual spins, thereby permitting calculations that exploit quantum interference effects within the computer itself [91].

2.2. *Magnetoelectronics*

During the past few years, quantum electronics and micromagnetics have begun converging towards a new field known as magnetoelectronics or ‘spintronics’, which focuses on low-dimensional electronic systems that display magnetically driven, spin-dependent phenomena [16]. On the one hand, quantum electronics has successfully exploited nanofabrication techniques to establish quantized energy levels, and charge manipulation between states produces a variety of electronic and optical devices. In parallel, research in micromagnetism has used miniaturization to produce submicron ferromagnetic structures whose switching, stability, and transport properties are controlled to create magnetic-storage and reading media. The merging of these two areas of research is giving rise to magneto-electronic phenomena where the spin of quantum-confined charge carriers is controlled using local magnetic fields. While there are several recent examples of systems where magnetic nanostructures are used in conjunction with semiconductor and metallic heterostructures to direct electron flow, the integration of magnetic and electronic quantum structures also offers opportunities to probe qualitatively new physics by obtaining additional dynamical information about the full quantum-mechanical nature of electronic states in reduced geometries. A fundamental understanding of incoherent and coherent electronic processes in nanometer-scale geometries will play an important role in the development of future technologies that rely on the quantum-mechanical control of electronic states. The eventual goal of such studies is to establish, store, and manipulate the coherence of electronic and magnetic spins in solid-state systems [92]. In order to attain this level of control, it is critical to develop an understanding of the microscopic mechanisms underlying spin injection, transport, and collection in practical material systems.

2.2.1. *Spin injection and transport*

Since the early part of this decade magnetoresistive films have served as read transducers to provide higher performance than their inductive coil predecessors in magnetic-recording technology.

However, the rapid pace of the disk drive industry, with magnetic bit areal densities increasing at a compound growth rate of about 60% per year, has already outdated these traditional MR sensors which cannot provide enough read signal for areal densities greater than ~ 5 Gbit/in². Currently, the disk drive industry is undergoing a revolution with the rapid introduction of novel GMR sensors, which can provide orders of magnitude more signal than conventional MR sensors. While the promise of GMR is high there are significant risks involved with the introduction of such a major new technology. Of particular concern is the long-term magnetic and structural stability of these novel structures within the relatively harsh environment of a disk drive enclosure.

The flux from the transitions between magnetic bits in conventional magnetic recording disk drives gives rise to a field at the read sensor of the order of a few tens of Oersteds. Thus, even though GMR multilayers, such as those formed from alternating layers of Co and Cu, each a few atomic layers thick, can exhibit MR values at room temperature exceeding 70% [93], these structures cannot be used for disk drive MR read sensors. Such multilayers require large magnetic fields of order ~ 10 –30 kOe to saturate their resistance because the interlayer antiferromagnetic exchange coupling provided by the ultrathin Cu layers is very large. Whilst it is possible to reduce the saturation field by using thicker Cu layers, it is preferable to use a spin-valve GMR structure which exhibits still significant changes in resistance ($\sim 10\%$) but at magnetic fields many orders of magnitude lower than GMR multilayered structures. The simplest spin valve structure comprises a soft ferromagnetic layer separated from a second harder ferromagnetic layer by a metallic spacer layer, typically copper. The resistance of the structure is changed when the magnetization of the soft layer is affected by external fields such as the flux from transitions in a magnetic recording medium (see Fig. 12). The detailed origin of the GMR effect in such structures has been the subject of much debate. An important question concerning the origin of the spin-dependent scattering processes which leads to GMR is now resolved in favor of a dominant contribution arising from spin-dependent scattering at the

interfaces between the ferromagnetic and metallic spacer layers [94]. The most sensitive spin valve MR sensors take advantage of this perhaps surprising result by doping the interfaces with thin magnetic layers, just a few atomic layers thick, which give rise to strong spin-dependent scattering. It is not surprising that the GMR exhibited by a spin-valve is very sensitive to details of its structure, which can be strongly influenced by the deposition method used to form the structure, and by the processing techniques used to define the MR sensor itself.

A second structure similar in construction to the spin-valve but whose MR arises from a quite different mechanism is the magnetic tunnel junction (MTJ). In an MTJ a thin insulating layer through which the electrons tunnel separates two ferromagnetic layers: the tunneling conductance of the junction depends on the relative orientation of the two ferromagnetic layers. MTJ, a curiosity for more than 20 years since its first discovery in 1977 [95,96] has become the focus of intense interest in recent years since large room temperature MR effects were found in such structures ranging from 10–18% [97,98] to more than 40% [99]. The mechanism of MR in an MTJ clearly is related to the extent to which the tunneling current is spin polarized. However, there are many remaining puzzles including, for example, the sign of the spin polarization of the tunneling electrons, and the relationship of the magnitude of the spin polarization to the magnetization and the detailed electronic structure of the ferromagnetic electrodes. Just as for GMR, the details of the structure of the interfaces between the ferromagnetic electrodes and, in this case, the tunnel barrier, are critical to determining not only the magnitude of the MR but also the bias voltage and temperature dependence of the MR.

Another important potential application of both GMR and MTJ structures is the use of such structures to form non-volatile magnetic memory cells (see Fig. 3b). For this application the MTJ or spin-valve devices are engineered to have two stable magnetic states in small magnetic fields. Typically, these states will correspond to parallel or anti-parallel alignment of the ferromagnetic layers in the sandwich. Note that a spin-valve MR read

head sensor is, by contrast, designed to have a nonhysteretic magnetic structure which responds approximately linearly to small applied fields. Understanding and controlling the magnetic response of these devices in both the spatial and temporal domains is a formidable challenge, particularly as the size of such devices decreases to deep sub-micron dimensions. The science of spin-dependent transport in GMR and MTJ structures is fascinating but progress in the detailed understanding of these phenomena, and in their technological applications, depends on a detailed characterization of their physical, electronic and magnetic structure both in unpatterned and in patterned structures.

The recent developments in GMR and MTJ materials has led to intensified interest in the possibility of building useful nonvolatile magnetic random access memory (MRAM). While there is strong interest in such nonvolatile magnetic memories for defense and aerospace applications, there is also great commercial potential for a nonvolatile MRAM technology that can compete, in performance, with existing memory technologies. In 1995, the defense advanced research projects agency (DARPA) funded three industry-led consortia to develop competing magnetic-memory technologies. Of these consortia, those led by Honeywell and Motorola have proposed using GMR elements and that led by IBM has proposed using memory elements comprised of magnetic tunnel junctions. While the latter give much higher magnetoresistance values ($>40\%$ at room temperature) than GMR structures of similar complexity, a concern is the control and reliability of the very thin tunnel barrier layers required. The success of any of these consortia depends on developing a magnetic memory technology that has excellent properties (short read and write times, low power, integration with standard complementary metal-oxide-semiconductor (CMOS) electronics) and scalability to ultra-high densities. For both GMR and MTJ elements, controlling the structure, especially the size and shape, and the micromagnetics of sub-micron-sized elements are critical issues that advanced microscopy techniques, now under development at the ALS, should help to address. Similarly, the thermal stability of these elements and their compatibility with standard CMOS processes is important. As

discussed above the interfaces between the ferromagnetic and the nonferromagnetic spacer layers (metallic for GMR and insulating for magnetic tunnel junctions) are critical to the properties exhibited by these devices. Advanced element-specific characterization techniques at the ALS should be helpful in understanding materials, processing, and reliability issues.

2.2.2. *Quantum confinement*

Quantum confinement of electrons takes on the added dimension of spin in magnetic nanostructures. Different spins experience different potentials and hence different degrees of confinement. Dimensionality is again important, and many types of structures and confinement can be envisaged. The magnetoresistive and interlayer coupling properties of layered GMR structures are one clear example of this type.

One of the last open basic questions in the physics of the GMR exhibited by layered structures is understanding the origin of the magnetic coupling in terms of momentum-resolved electronic structure. In particular, the ‘long-period’ oscillation of iron/chromium layers remains a controversial issue with multiple contradictory theoretical predictions [100,101] and partial experimental findings [102]. Angle-resolved photoemission studies to monitor the emergence of spin-polarized quantum-well states across the Fermi energy as a function of the thickness of the spacer medium (e.g., chromium) are needed to solve this problem. Many transition-metal spacers (chromium, niobium, molybdenum) yield provocative predictions, and the band structure of transition metals yields Fermi surfaces that are complex and rich in possibilities. It is thus a major challenge to move beyond understanding the electronic structural origins of the couplings in noble metals (copper, silver, gold) to these more complicated structures that will require experimental probes capable of momentum and spin-resolved detection of electrons involved in the quantum-well states at surfaces. Probing spin-polarized quantum-well states in buried layers poses even more experimental challenges. And finally, being able to more directly probe the degree of interface mixing and the atomic and magnetic structures of atoms near the interfaces that are

crucial to the GMR effect (as well as other magnetic nanostructures) is a key area of future experimental need.

2.2.3. *Magnetic semiconductors*

A very appealing class of materials with unexplored spin dynamics is provided by semiconductor nanostructures doped with magnetic moments [103]. These systems provide a unique nexus between low-dimensional magnetism and semiconductor quantum confinement. By tailoring the quantum-confining potential, one can systematically control the overlap between quantized electronic states and the local moments, while the magnetic environment generated by the local moments may be varied through factors such as strain, dilution, and dimensionality. The exchange interaction between the electronic band states and the local moments leads to a greatly enhanced spin splitting of the confined carriers in the presence of an applied magnetic field, resulting in spin-dependent and magnetically tunable confining potentials. Since the various interactions involved (e.g., the d–d and sp–d exchange) are well characterized because of the extensive work in both bulk magnetic-semiconductor crystals and magnetic-semiconductor heterostructures, these magnetic nanostructures provide clean model systems for basic studies of electron-spin dynamics in low dimensions. Furthermore, these materials have excellent optical properties that are characterized by small inhomogeneous line widths and well-defined excitonic resonances. Hence, the development of powerful probes of spin dynamics and magnetic interactions has been possible recently using state-of-the-art femtosecond-laser magnetooptical techniques [53].

Within this field during the past few years, there have been remarkable advances in materials science that are fueling a resurgence of activity. Recently, it has been shown that one may fabricate ‘digital magnetic heterostructures’ in which interactions between localized magnetic spins and their wavefunction-overlap with quantized electronic states is tuned through a controlled distribution of two-dimensional magnetic layers [104]. These engineered planar structures reduce clustering of the magnetic moments, thereby resulting in an

enhanced paramagnetism, allow a large magneto-electronic overlap, and display qualitatively and quantitatively different dynamical interactions as compared to bulk alloys. In addition, it has now become possible to electronically dope these structures with either n-type or p-type dopants, representing a marked advance in potential applications of these materials. As in traditional semiconductor physics, the formation of a two-dimensional electron gas in modulation-doped nanostructures has proven to be a mainstay of contemporary interests in both condensed matter physics and quantum device physics. The recent fabrication of magnetic two-dimensional electron gases has opened a new model system, in which a two-dimensional population of electrons interacts ferromagnetically with local moments, in materials readily accessible to quantum transport and magnetooptical studies [105]. Measurements have shown that such a magnetic two-dimensional electron gas is easily spin-polarized in modest applied fields, even at large filling fractions, since the spin splitting at cryogenic temperatures is much greater than the Landau-level splitting. Furthermore, the last few years have seen the creation of semiconductor nanostructures with dimensions less than two. Zero-dimensional quantum dots have attracted substantial attention because of recent successes with in situ fabrication of defect-free, self-assembled ensembles of strained semiconductor quantum dots with surprisingly high quantum efficiencies and relatively narrow size distributions [106].

These scientific opportunities in magnetic semiconductors have helped stimulate new fabrication and nonequilibrium growth techniques aimed at producing truly integrated magnetoelectronics, with nothing short of spectacular results. A number of remarkable discoveries suggest that detailed structural studies may have a profound impact in this area. For example, it has been historically difficult to produce magnetic semiconductors using technologically common semiconductors; moreover, the introduction of magnetic moments, in these materials has typically resulted in Heisenberg antiferromagnetic interactions, thus limiting these systems to behaving as heavily diluted paramagnetic media. However, recently it has been demonstrated that nonequilibrium molecular-

beam-epitaxy (MBE) growth techniques may be used to incorporate Mn^{2+} ions substitutionally into GaAs semiconductors, where they also act as acceptors or p-type dopants. In contrast to all theoretical predictions, this combination of events has produced the first ferromagnetic semiconductor grown by MBE that may be used to construct nanometer-scale ferromagnetic devices [107]. This revolutionary discovery has rapidly led to demonstrations of spin-polarized resonant-tunneling diodes and the genuine possibility of a new family of spin-injection devices based on semiconductor technology. The advent of highly magnetoresistive ferromagnetic materials that are compatible with the semiconductor industry has profound economic implications. There is a great deal of fascinating scientific work to do in understanding the mechanism driving ferromagnetism within a semiconductor host and understanding in general how the electronic structure of wide-gap semiconductors may be tuned to modify exchange interactions in solids. This knowledge may subsequently be used to generate new families of artificially engineered band gap systems that incorporate ferromagnetic media for science and technology.

2.2.4. Disorder

The effects of disorder and defects on transport properties in conventional metals and semiconductors are quite important in practice and hence fairly well established. The effects of magnetic disorder on spin-dependent transport are less well understood, although likely to be just as important in magnetoelectronic applications and more complex in nature. Disorder can take many forms in magnetic materials, including the usual chemical and structural disorder but now with local magnetic properties varying site by site [108,109]. Simple examples of this magnetic disorder are nanoscale chemical/magnetic clustering in metallic alloys that introduce spin-dependent scattering centers and increase magnetoresistance over that of a nominally homogeneous alloy. Interfaces and domain walls are themselves forms of disorder that can significantly influence magnetotransport, as mentioned above [110,111].

Recently, it has become clear that the presence of disorder may be beneficial in certain classes of

materials. For example, the presence of manganese ions introduced through nonequilibrium-growth techniques into the III–V semiconductor GaAs has produced ferromagnetic GaMnAs. In this example, the addition of magnetic moments into the lattice results in a ferromagnetic moment, p-type doping, and potential disorder. The combination of these latter two attributes result in a new band structure that generates a long-range spin interaction and subsequent ferromagnetism in a MBE-grown semiconductor host. While the applications of these new materials are numerous, ranging from magnetically tunable resonant tunneling diodes to magnetic field-effect transistors, the fundamental mechanism for ferromagnetic exchange remains poorly understood. Resonant tunneling diodes based on GaAs/ErAs, where magnetization-controlled quantum transport has been demonstrated in a materials system with highly mismatched interfaces and dislocations, are another outstanding example [112]. In addition, strong optically active materials, such as GaN, have five to six orders of magnitude higher defect density than today's commercial semiconductors, yet are amongst the most promising materials for blue lasers and display technologies [113]. It is expected that the role of disorder and its effect on micromagnetics and new hybrid magnetic-semiconductor materials may be uniquely explored with synchrotron radiation.

2.3. Structure and magnetic order

Textbooks illustrate the interdependence of interatomic structure, electronic structure, and magnetic order in the bulk of prototypical ionic and itinerant ferromagnets, ferrimagnets, and antiferromagnets. The foundations provided by models of these prototypical systems provide starting points for understanding the increasingly complex materials of current and likely future interest. Experimental techniques capable of providing element-resolved information about local structure, electronic structure, magnetic moments, and magnetic structure at relevant length scales (e.g., short-range order versus long-range order over a continuum of lengths) are essential for future studies of magnetic materials.

2.3.1. Magnetic anisotropy

As discussed in Section 2.1.1, magnetic anisotropy is linked to structural anisotropy, and the mechanisms linking structural to magnetic anisotropies can differ from material to material. Small strains, surface or interface effects, texturing, and growth in applied fields are examples of effects known to influence anisotropies in specific materials.

2.3.2. Frustration

The role of structure in quenching moments and frustrating spin alignments has been well studied in many bulk materials, and now it must be extended into magnetic nanostructures where the effects of surfaces and interfaces must be understood. An interesting example is the interface between ferromagnets and antiferromagnets involved in exchange biasing [68,69], as discussed in Section 2.1.1 and used in some GMR structures [114] such as the prototypical iron/chromium multilayer system. In each case, the issues revolve around spin frustration in the interfacial region. In the iron/chromium case, it is also of interest to understand how the spin-density waves in the chromium are affected by the confined geometries and how placement of nodes in the spin-density waves in these regions may minimize the frustration energy by limiting the magnitude of the chromium moments in the interfacial region [115]. Neutron scattering has served as a prime tool to explore the interfacial physics in these structures [116], but VUV/soft X-ray scattering and spectroscopic measurements with third-generation synchrotron radiation should provide additional information. The physics of noncollinear spin structures in magnetic heterostructures represents a general area of ongoing research that is rich in opportunities to be explored by element-specific, spin-polarized spectroscopies and spectromicroscopies.

2.3.3. Proximity effects/induced magnetism

Many basic- and applied-research problems require the use of magnetic materials in contact with other magnetic, nonmagnetic, superconducting, semiconducting, or insulating materials. By analogy with the superconducting proximity effect, it is natural to question if there exists a magnetic

proximity effect whereby the magnetic properties of one material influence the properties of the other [117–119]. Many different issues must be distinguished in considering this question. For example, interdiffusion, reaction, and roughness at interfaces can have important physical consequences, some of which might include induced moments in nominally nonmagnetic atoms through hybridization of electronic states [120]. Such induced moments have been observed with element-specific synchrotron techniques and can be thought of as one type of proximity effect mediated by overlap of electronic states [37–39]. Such induced moments raise interesting questions about itinerant magnetism but are distinct from a more general notion of proximity effects. Thus, it is important to understand whether the magnetism in one material can create and affect magnetism in another material that is in close physical proximity. Simple mean-field theory predicts the presence of this type of effect in any system undergoing a second-order phase transition and was the subject of some theoretical research 15–20 years ago. Layered magnetic systems are again candidates for the investigation of such effects, which in addition require sensitive experimental probes capable of isolating the magnetic responses of individual layers and constituent elements. Indeed, element-specific XMCD has recently been used to study the effect on the Curie temperature of two different magnetic layers separated by an ultra-thin nonmagnetic layer [121]. It would be interesting to investigate the dependence of proximity effects on their coherence lengths, temperature, and other experimental parameters. Such effects would clearly impact the study of all magnetic nanostructures, and they could be particularly useful in device applications if they could be manipulated or controlled.

2.3.4. *Interfacial effects*

With the rapid trend towards ever-smaller magnetic nanostructures, the characterization of the interfaces inherent in these structures becomes increasingly important [122]. Real interfaces can always exhibit imperfections and roughness, even if there are examples of atomically smooth interfaces between certain pairs of materials as grown in carefully controlled, often UHV, environments. Rough-

ness can be categorized in many ways, but one useful classification is based on spatial frequencies. Roughness at the highest (atomic-scale) frequencies corresponds to intermixing between atoms at the interface; it could result from growth-induced (knock-on) mixing, interdiffusion during or after growth, or chemical driving forces leading to compound formation at the interface. At progressively lower spatial frequencies, roughness can be described as topological and characterized by height variations of a distinct interface and their correlation with in-plane distance. A variety of magnetic phenomena are affected by interfacial roughness, and indeed magnetic roughness can be defined as the variation in magnetic properties over this same range of spatial frequencies. Early neutron scattering [108,109], and recent soft X-ray magnetic-scattering measurements [46,48], suggest that chemical and magnetic roughness at interfaces are not necessarily the same. The characterization of these various types of roughness at real (buried) interfaces is challenging, and it will become increasingly important both for fundamental understanding and because these imperfections may directly influence magnetotransport and other properties of interest in device applications. Techniques capable of quantifying magnetic roughness and magnetic correlations in heteromagnetic structures will therefore likewise become increasingly important.

In the areas of surface, monolayer, and thin-film magnetism, the last decade was driven by the quest to verify theoretical predictions of enhanced magnetic moments [123,124], reduced or increased magnetic transition temperatures, and enhanced and/or perpendicular anisotropies. We anticipate that the next decade will involve further explorations of the role of unique magnetic surface and interface states in underlying the phenomenon of surface-altered magnetic order. Exchange biasing of a ferromagnetic layer by an antiferromagnetic layer is one area where interfacial magnetic states appear to control important properties [125]. Fundamental issues will also include the elucidation in model systems of the collapse of the exchange splitting above the Curie transition in itinerant ferromagnets as a function of k -vector. Experimental techniques capable of providing spin-polarized electronic structural information, as well as

geometrical structural information, are valuable in this area. The surface magnetooptic Kerr effect, valence and core photoemission, X-ray absorption, X-ray emission, and neutron scattering have provided valuable input to date in these areas, and we expect future developments to enhance most of these techniques.

2.4. Exploratory materials

In this section, we consider some novel magnetic materials that are currently beginning to be explored. Although perhaps not immediately amenable to technological application, these materials provide new and exciting ways to look at magnetism in the future, as well as additional fundamental questions to be explored.

2.4.1. Hybrid structures/competing interactions/frustrations

All of the magnetic nanostructures discussed above can be thought of as hybrid structures. Structures containing normal-metal, antiferromagnetic-metal, and insulating-spacer layers have already been mentioned. Superconducting interlayers also suggest new concepts for spin-injection, all-metallic spin-transistors, and novel magnetoresistive memory elements. For example, a new type of superconductive coupling across ferromagnetic layers involves a change in the phase of the order parameter by 180° (π) across the ferromagnet [126,127]. The phase shift oscillates, according to theoretical prediction, as a function of the thickness of the ferromagnetic layer, from its ordinary value of 0 to its ‘exotic’ value of π , and physical properties, such as the superconductive transition temperature, critical field, and critical current are all predicted to be nonmonotonic functions of the thickness of the ferromagnetic spacer [128]. This ‘ π -phase’ superconductivity is a challenge to create in the laboratory and to characterize with credibility [129]. All of the tools of precision growth, as well as interfacial characterization, come into play here.

The converse problem of interleaving ferromagnets and superconductors in order to perturb magnetism (rather than superconductivity) offers unusual challenges as well [130–134]. It is well known that magnetism and superconductivity

represent competing interactions that tend to preclude each other. This is because spin breaks the time reversal symmetry of the Cooper pairs ($+k\uparrow$, $-k\downarrow$). Thus, by extending the recent dramatic advances in the use of photoemission to observe the superconducting energy gap and its anisotropy in exotic high- T_c materials, the possibility exists to observe the superconducting/ferromagnetic interface and characterize the *breaking* of Cooper pairs and the introduction of states and even impurity bands into the superconducting energy gap [135,136]. While such observations are possible in tunnel junctions [135,136], the k -space origin of the states and the anisotropies can be explored uniquely via photoelectron-spectroscopy techniques. This capability can add to our fundamental knowledge base and might help formulate new paradigms that challenge the quasiparticle concept.

2.4.2. Active interfaces

Mixing and matching ‘active’ materials like superconductors and ferromagnets with semiconductors will produce material systems with new properties important for future electronics, photonics, and magnetics. The important materials, physics and device issues center upon: (1) the growth and character of the interface between wildly dissimilar materials, (2) understanding phase-coherent or spin transport across the interface, and (3) the interaction between nanostructured ‘active’ materials each in intimate contact with high-mobility semiconductor structures. The electronic nature of the interface typically determines the behavior of the composite system. For example, although there has been great progress in understanding the coherent transport across the Nb-InAs superconducting-semiconducting interface, control of the quality of that interface remains the most critical issue in reproducible preparation of systems that demonstrate phase-coherent transport [137]. Less well developed but scientifically and technically important are ferromagnet-semiconductor heterostructures, where the order parameter in the contact is the magnetization, and the critical issue is the transport of spin polarization across the interface and through a two-dimensional electron gas.

A prototypical active interface may be viewed as a semiconductor quantum heterostructure in direct contact with a ferromagnetic film. For example, one may consider the structure consisting of a resonant-tunneling device, where the local magnetic fields will have a dramatic effect on the electronic behavior of the structure through proximity effects. This class of systems offers the potential of exploiting ultrafast quantum electronics to sense magnetic switching, thereby giving rise to another family of composite magnetoelectronic materials. The nature of the ferromagnetic–semiconductor interface, local band structure, and chemical potentials must be understood and engineered in order to produce these systems. Synchrotron radiation will play an important role in determining the character of such interfaces in concert with in situ growth characterization and modeling.

2.4.3. Biomagnets

An ordered magnetic microstructure may be fabricated by either a top-down approach by depositing bulk material and using standard lithographic techniques or a bottom-up approach by assembling nanometer-scale ferromagnets created by chemical synthesis. The first method allows direct spatial control and can produce individual magnets as small as 10–100 nm using electron-beam lithography or local deposition with a scanning tunneling microscope. The second approach includes magnetic clusters and implanted ion species. Although there is little spatial control in the chemical synthesis beyond direct masking, they can produce structures of smaller dimensions than lithography. Another intriguing possibility is to use biological systems to direct the self-assembly of ordered magnetic structures. For example, certain organisms, such as magnetotactic bacteria, assimilate magnetic materials and create microscopic ferromagnets of remarkably high purity and uniformity [138]. Others create complex inorganic structures through biomineralization, an approach that may be used as a template for artificial magnetic structures [139]. Thus, bacteria-based systems are now being used as frameworks for ferromagnetic microstructures, such as bacterial threads, typically of order 100 μm in diameter and several tens of centimeters long, that are fabricated by pulling many

strings of cells together out of a culture solution [140]. The resulting ‘bionites’ have high densities of particles about 100 nm in size and extremely high tensile strengths. Biomaterials-engineering techniques, such as bacterial templating, are likely to play an increasing role in the development of future technologies, with the potential of providing ordered arrangements of microscopic particles over macroscopic length scales. Chemical specificity for nucleating magnetic entities within these complexes require atomic-scale characterization techniques that may be addressed by synchrotron radiation.

2.4.4. Molecular magnets

Remarkable new techniques in molecular chemistry are now making it possible to create true ‘molecular magnets’, in which the magnetic ions are added one at a time and the resulting magnet has a precisely defined atomic weight [141]. Many of these new magnetic molecules are constructed such that the magnetic cluster is secluded from contact with its external environment by a shell of organic ligands. For example, one such magnet, ‘Fe10’, is dubbed the ‘ferric wheel’ by its inventors because of the ring structure formed by the ten iron atoms and the bridging organic anions. Magnetization measurements reveal that this particular cluster is literally a perfect molecular antiferromagnet with an $S = 0$ quantum ground state [142]. The successive transitions from one discrete quantum spin state to the next, evident for magnetic fields up to 45 T, resemble a sequence of phase transitions in the magnetic order of bulk layered antiferromagnets. The ability to fabricate macroscopic single crystals of these materials containing perfectly spaced arrays of identical molecules offers the promise of fascinating new science with potential applications to technology. This is thus magnetoelectronics at the most basic level. These molecular magnets may be used to experimentally bridge the gap in our understanding of micromagnetics from the atomic to the mesoscopic length scale, and they have already been shown to be nearly ideal systems in which to explore the macroscopic quantum tunneling of magnetization [143] (as discussed in Section 2.1.6 for more complex systems).

3. VUV/soft X-ray capabilities

Valence and core-level spectroscopies in the VUV/soft X-ray region couple directly to the electronic states responsible for magnetism in metals, insulators, and semiconductors, and so they are immensely relevant in the study of magnetism and magnetic materials. VUV radiation in the 10–50 eV spectral range provides optimum access to valence levels and hence the electronic spin structure of all materials. The spectral range from 50–2000 eV is of prime importance for core-level photoemission and high-resolution X-ray absorption, emission, and scattering spectroscopies. In this range, an abundance of core levels exist from which electric dipole transitions couple spin–orbit split initial states to spin-polarized final states. Long core hole lifetimes result in strong, sharp lines in the photoelectric cross section and spectacular resonances in refractive properties that together give the optical response of materials. Of particular importance for magnetism are the 2p core levels (L edges) of the magnetic 3d transition metals (chromium, manganese, iron, cobalt and nickel), which lie in the 550–900 eV range, and the 3d core levels (M edges) of the rare earths, which lie in the 800–1600 eV range. In X-ray absorption and resonant scattering, electric dipole transitions from these core levels allow direct access to the magnetic properties of the important 3d valence shell of the transition metals and the 4f valence shell of the rare earths. Core-level photoemission from the 3s, 3p, and 2p levels in the 3d elements [40,42,43,144,145], and 4p and 4d levels in the rare earths [44] also permits studying magnetic properties through multiplet effects and magnetic dichroism. Other core levels of interest include those of the chemically important elements boron, carbon, nitrogen, and oxygen, which have 1s levels (K edges in the 150–550 eV range and are found frequently in interesting materials. In general, absorption and resonant-scattering (including reflectivity) spectroscopies directly probe the empty states at and above the Fermi level or band gaps, while valence photoemission and X-ray emission spectroscopies directly probe the filled electronic states. Resonant inelastic or Raman scattering may provide sensitivity to magnetic excitations. Some general capabilities of using VUV/soft X-ray

probes for the study of magnetic materials are enumerated in Table 2.

Spin-polarized photoemission [146] is well-established as the premier tool for the determination of the spin-dependent electronic structure. Because of factors such as cross-section effects, energy resolution, and momentum resolution, the spin-dependent electronic valence-band structure is best studied by use of VUV radiation in the 10–50 eV range. At higher photon energies spin-resolved core-level photoemission [40–44], opens up the study of element- and chemical-state-specific magnetic properties. Chemical-state and site information is present in core-binding-energy shifts; for example, it has been possible at the ALS to resolve the atoms at a nonmagnetic/ferromagnetic interface and to study their local atomic structure via site-specific photoelectron diffraction [147]. In addition, photoelectrons excited from the core levels contain magnetic information through multiplet core-valence coupling in the emitting atom or through spin polarization imparted by excitation of a spin–orbit-split level with polarized radiation. This can lead to exchange scattering of the photoelectrons by magnetic neighbor atoms. The later effect has been termed spin-polarized photoelectron diffraction, and it provides information on the short-range spin structure around a given type of emitting atom in the sample. It thus directly determines the local magnetic structure and is

Table 2

Capabilities of VUV/soft X-ray techniques in the study of magnetism and magnetic materials

General capabilities of VUV/soft X-ray synchrotron techniques

Spin-dependent electronic structure
Elemental specificity
Chemical specificity
Sensitivity to local magnetic structure
Angular momentum specificity
Sensitivity to spin and orbital moment
Sensitivity to ferro-, ferri-, and antiferromagnetic alignments
Bulk and surface sensitivity

Unique capabilities of high-brightness synchrotron facilities

Magnetic spectromicroscopy
Enhanced capabilities for all spectroscopies and scattering experiments, including spin and time resolution

therefore applicable to systems without long-range geometric order. To date, this technique has been applied to both antiferromagnets [148–150] and ferromagnets [151–153]. It has also been proposed to carry out spin-polarized photoelectron holography [147] by measuring the differences in photoelectron diffraction patterns for spin-up and spin-down electrons (e.g., from multiplets). This technique would permit directly imaging magnetic spin moments in space at atomic resolution, and it represents an interesting future experiment whose experimental difficulties would make it impossible to perform without a high-brightness third-generation source.

The natural polarization of synchrotron radiation enables all magneto-optical effects (magnetic circular and linear dichroism, Faraday and Kerr magneto-optical rotation, etc.) common in the near-visible spectral regions to be extended into the soft X-ray range, where they gain element specificity when applied in spectroscopic fashion near core-level absorption edges. X-ray magnetic circular dichroism is very commonly used today to measure the difference in absorption of circular polarization with opposite helicity. Faraday and Kerr magneto-optical-rotation signals result from the difference in refraction of circular polarization components with opposite helicity, and are measured using tunable linear polarizers [49,50]. The Faraday magneto-optical-rotation spectrum is related to the XMCD via a Kramers–Kronig dispersion transformation. Scattering techniques relying on intensity measurements generally measure a signal influenced by both absorptive and refractive magneto-optical responses.

Powerful sum rules directly link XMCD spectra to the value of the atomic spin and orbital moments [32,33] and their anisotropies [34]. The direct separation and determination of atomic spin and orbital moments is a unique capability of the core-level magneto-optical spectroscopies. Like core-level photoemission, such measurements are element specific and, through additional near-edge fine structure, also provide sensitivity to the local bonding environment of a given atom [154,155]. Because the allowed core-to-valence transition is dictated by the dipole selection rule (i.e., $s \rightarrow p$, $p \rightarrow d$, $d \rightarrow f$), excitation at different absorption

edges provides selectivity to the angular momentum of the valence-electron states. For example, L-edge (2p) excitation in cobalt probes the cobalt 3d valence subshell while K-edge (1s) excitation probes the cobalt 3p valence electrons [38,39]. The large size of the circular dichroism effects (up to 30% in XMCD and tens of degrees in magneto-optical rotation) allow the study of very dilute and/or weak moments. Taken together, these general capabilities of X-ray magneto-optical spectroscopies provide powerful tools to dissect the net magnetic behavior of complex samples (both homogeneous and heterogeneous) into the contributions from the atomic and chemical constituents.

Core-level dichroism in X-ray absorption and photoemission also allows the determination of the magnetic axis and the study of magnetic correlations in antiferromagnets. Such systems are of great importance, but their study is often impeded by their compensated overall moment. For example, linearly-polarized X-ray absorption spectroscopy can sense the orientation of the magnetic axis relative to the orientation of the electric field vector [156–160]. In the presence of a magnetic axis, the spin–orbit coupling in the valence shell leads to a slight anisotropy of the valence charge, which can be detected by polarized X-ray absorption. Temperature-dependent measurements further allow the separation of the magnetic effect from a valence-charge anisotropy caused by the electrostatic crystal potential. The linear magnetic dichroism is particularly large when the X-ray absorption spectrum exhibits sizeable multiplet structure. In addition, multiplet effects in core photoemission can be used to probe short-range magnetic order in antiferromagnets (e.g., using spin-polarized photoelectron diffraction), as each emitter in this case acts as its own spin reference.

The buried layers and interfaces often critical in real magnetic nanostructures can be uniquely studied with soft X-ray techniques. Photoemission or X-ray absorption techniques relying on electron detection are typically surface or near-surface sensitive, with the information depth given by the escape depth of the particular photoelectrons or secondary electrons detected. In typical materials for magnetic applications, these depths range from a minimum of approximately 5 Å for photoelectrons at about

50 eV to 15 to 20 Å for photoelectrons at 1000 eV. In soft X-ray absorption measurements, the electron-yield sampling depth is determined primarily by secondary electrons, and it varies from 20 to 40 Å for metals to 50 to 100 Å for insulators. Emerging techniques relying on photon detection can have much greater information depths ranging from 0.1 to 1 µm at energies away from any absorption edges to a few tens of Angstroms at edges and in a total-reflection geometry. Working with electron detection at either much lower or much higher energies is another avenue for increasing bulk sensitivity in even these measurements. Thus, surface and bulk sensitivity can be obtained by choosing the detected particle and the relevant energies, and with careful attention to the relevant penetration and escape depths, it is possible to vary the sensitivity from the first few atomic layers to depths throughout the bulk (a true bulk measurement), thus isolating signals associated with buried layers or interfaces. Photon-based techniques can also be applied in strongly varying fields, thereby allowing element-specific hysteresis measurements of interest in understanding reversal behavior in complex systems [50,161]. With soft X-ray wavelengths ranging from 0.5 to 50 nm, scattering techniques offer interesting possibilities to study magnetic correlation lengths on the nanometer scale and above, and they do not require long-range order.

Many of the above capabilities have been pioneered at existing synchrotron-radiation facilities, and hence they do not require the brightness of third-generation facilities for continued application. However, even for these capabilities, the increased brightness of third-generation sources will translate into new experimental opportunities, some of which can be extrapolated from existing work. For example, in photoemission-spectroscopy measurements, the increased brightness can translate directly into increased energy and k -space resolution and into much decreased data-collection times, particularly for spin-resolved measurements using inherently inefficient (10^{-4}) Mott or other detectors. Additional advantages in photoemission will derive from the ability to resolve core-level shifts, multiplet splittings, and spin-dependent scattering effects that would be prohibitively slow to measure with a second-generation source. In

photon-based measurements, increased brightness can enhance diffuse magnetic scattering measurements, and the transverse coherence of undulator sources offers the possibility of dynamic magnetic-speckle scattering. In general, the increased intensities associated with higher brightness will allow all types of experiments to move farther into the time-resolved domain, whether it be in situ studies of the growth of magnetic films or time-resolved studies of the dynamics of magnetization-reversal processes.

A key advantage of the ALS in magnetism research lies in its unique capabilities for magnetic spectromicroscopy that results from the higher brightness. Magnetic microscopy with soft X-rays offers several unique capabilities that are similar to those of X-ray linear- and circular-dichroism spectroscopies; i.e., high magnetic spatial resolution is complemented by elemental, chemical, and variable-depth sensitivity [162]. The latter ability, for example, allows one to image magnetization distributions separately in different magnetic layers of magnetic heterostructures. High spatial resolution is achieved by several means [162], three of which are illustrated in Fig. 5. The first is based on producing a focused X-ray beam by means of suitable X-ray optics, such as a zone plate, and creating a microscopic image of the sample by scanning it laterally relative to the focal spot. Either the transmitted X-ray intensity or a signal from the sample, such as the fluorescence or electron yield, can be detected, with different depth resolutions for each case. In the scanning technique, the resolution is determined by the size of the X-ray spot and today lies in the 30–40 nm range, but a resolution of 20 nm is expected in the near future. In the second method, a condenser optic such as a zone plate illuminates the object while an imaging zone plate produces an image on a detector. In the third method the sample is illuminated by a X-ray beam that is only moderately focused, e.g., to tens of micrometers, so that the spot size matches the maximum field of view of a photoelectron electron microscope (PEEM). The lateral resolution is determined by the electron optics in the PEEM and a resolution of 22 nm has already been obtained [163]. Designs for future spectromicroscopes indicate that a 2 nm resolution may be possible [164].

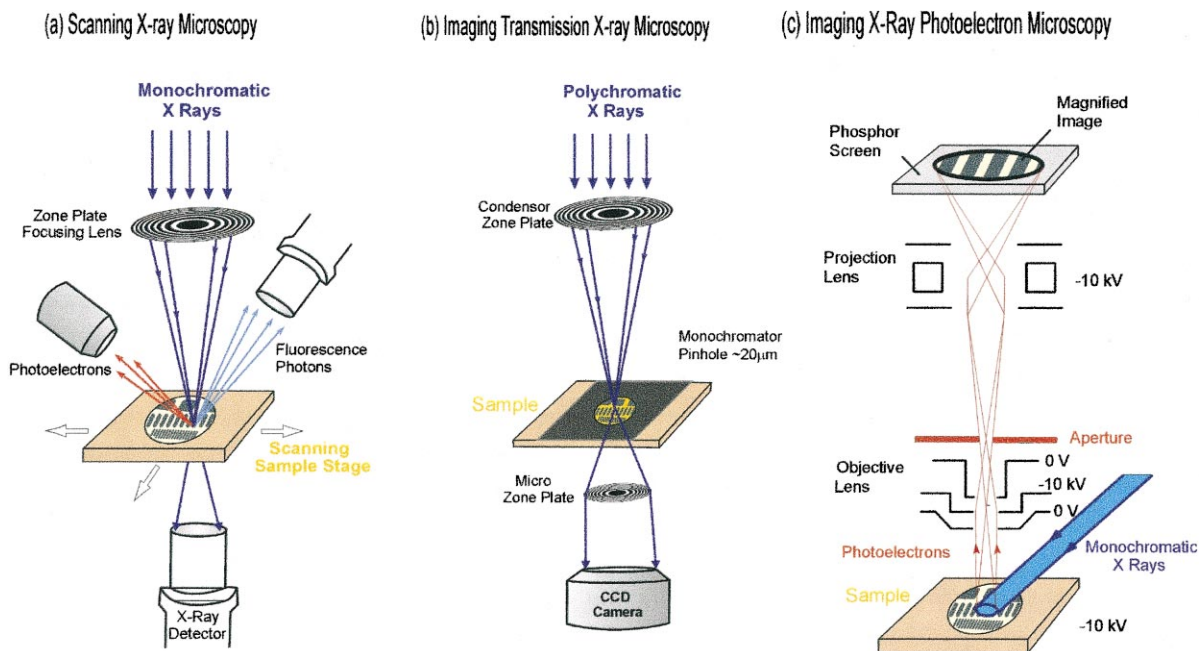


Fig. 5. Principles of scanning X-ray microscopy and two imaging X-ray microscopy techniques are shown. In the scanning mode (a) a small X-ray spot is formed by a suitable X-ray optic, e.g., a zone plate lens as shown, and the sample is scanned relative to the X-ray focal spot. The spatial resolution is determined by the spot size. The intensity of the transmitted X-rays or the fluorescence or electron yield from the sample is detected as a function of the sample position and thus determines the contrast in the image. In imaging transmission X-ray microscopy shown in (b), a condenser zone plate in conjunction with a pinhole before the sample produces a monochromatic photon spot on the sample. A micro-zone plate generates a magnified image of the sample that can be viewed in real time by a X-ray sensitive CCD camera. The spatial resolution is determined by the width of the outermost zones in the micro zone plate. In imaging X-ray photoelectron microscopy shown in (c), the X-rays are only moderately focused in order to match the field of view of an electron microscope. Electrons emitted from the sample are projected with magnification onto a phosphor screen and the image can be viewed in real time at video rates. The spatial resolution is determined by the electron optics within the microscope, the size of the aperture and the operation voltage (from Ref. [162]).

To date, magnetic images using soft X-rays have been obtained by imaging emitted secondary electrons [57], Auger electrons [165], or photoelectrons [163], and also in transmission using zone-plate optics and photon detection in imaging [166] and scanning [50] modes. Other forms of magnetic microscopy with soft X-rays, such as Kerr microscopy using photons in reflection, can also be developed.

4. Current and future science with VUV/soft X-rays

In this section, we discuss a few particularly stimulating examples of recent work in magnetism that have made use of VUV/soft X-ray radiation,

and other areas for fruitful future work at a third-generation source like the ALS.

4.1. Magnetic anisotropy

Core-level X-ray magnetic dichroism spectroscopy offers the unique capability of separating the spin from the orbital moment through sum-rule analysis. It is, therefore, ideally suited to study one of the fundamental issues in magnetism, the microscopic origin of the magnetocrystalline anisotropy, as discussed in Section 2.1.1. Recent angle-dependent XMCD studies in high magnetic fields have been used to directly link the thickness-dependent magnetic anisotropy in Au/Co/Au multilayers with

the preferred direction of the orbital moment [34,36], as shown in Fig. 6. These studies have provided a way to visualize the microscopic origin of the magnetocrystalline anisotropy in a simple picture based on moment anisotropy rather than the less intuitive concept of energy anisotropy. In this picture the anisotropy of the lattice or local environment causes a preferred direction of the orbital moment, which then redirects the isotropic spin moment into a parallel or antiparallel alignment (Hund's rule) by means of the spin-orbit coupling. Early work, which has addressed multilayer and thin-film systems where the magnetocrystalline anisotropy is large [36,167,168], points to many future opportunities. Improved measurement methods, e.g., transverse geometries [169] and standing-wave techniques, will extend these studies to samples with smaller anisotropies and allow detailed investigations of dimensionality and inhomogeneity effects with both lateral and depth resolution. The quantitative link between macroscopic measurements of anisotropy energy and the average moments obtained from these spectroscopies is one direction for future research [170].

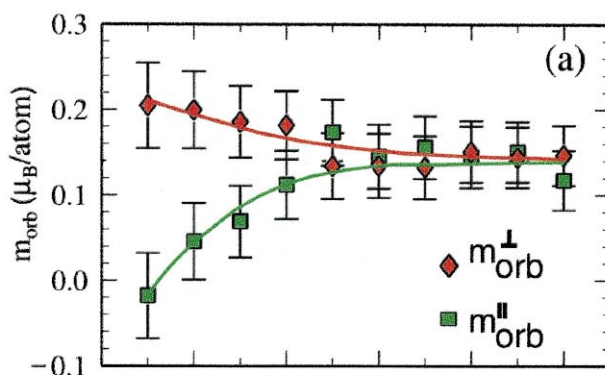
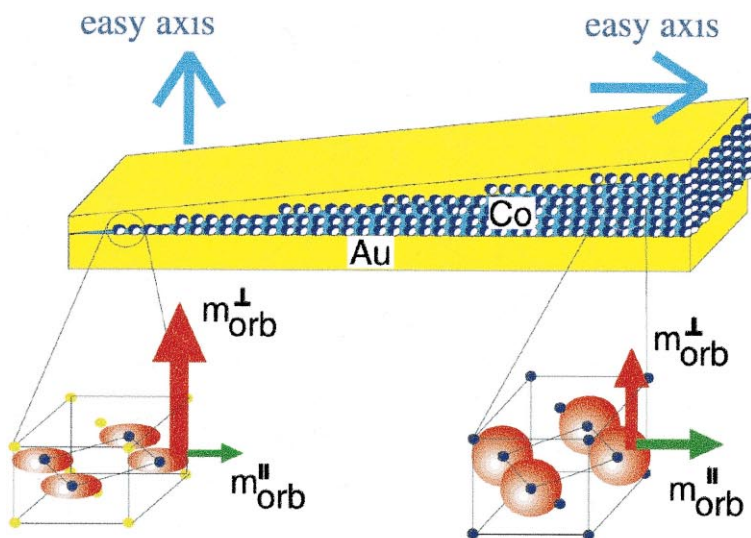
One example of a future application of X-ray capabilities is the study of spring magnets, as discussed in Section 2.1.1. Here, the elemental specificity of core-level magnetooptic techniques will allow the separation of the magnetic response of the soft and hard materials in the hybrid structure. Not only could the average spin and orbital moments on individual species in different layers be probed, but magneto-optical rotation and other methods could be utilized to measure the hysteresis response of the different layers individually to more fully understand magnetization reversal in these structures. Such studies should provide a better understanding of the unusual twist structure predicted in the soft layer, and they also should provide information concerning the interfacial region where little is known about how the transition from hard, high-anisotropy material to soft, low-anisotropy material is made.

4.2. *Structure and magnetic properties*

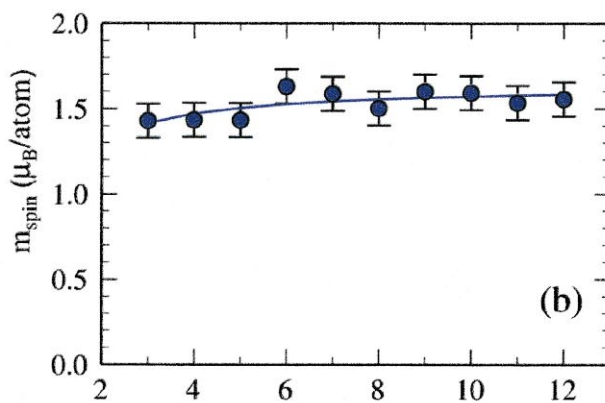
A key issue in nanoscale magnetics is the interrelation of structure and magnetic properties. Stud-

ies of surface and ultrathin-film magnetism are extending beyond elemental overlayers, such as epitaxial iron layers, to surface and thin-film alloy systems, where structural aspects of pseudomorphic or more complex growth modes can influence both structure and magnetism in overlayers. An example of the strong interplay of geometric structure and magnetic properties can be seen in Fig. 7, where Invar quenching is observed in NiFe ultrathin films despite the absence of the phase change from FCC to BCC that is observed in the bulk [171]. Here in the ultrathin film, the phase change is frustrated, and instead a decrease in the unit-cell volume is correlated with the loss of magnetization. In turn, understanding these NiFe ultrathin films has a technological significance because they are frequently used in GMR and spin-valve devices. Increased intensities resulting from higher brightness should enable in situ growth studies of layered structures, perhaps with dynamic sensitivity. Being able to probe the local atomic structure around each elemental constituent via photoelectron diffraction will also assist in characterizing such structures fully.

Probing magnetic structure both laterally and into the depth of thin film, layered and bulk structures is feasible using scattered or reflected photons. Resonant and nonresonant elastic and resonant inelastic scattering have been applied in the hard X-ray range to study a variety of fundamental aspects of magnetic structure [172–176]. At hard X-ray wavelengths these techniques often involve diffraction to study magnetic structure at interatomic distances. Similar techniques are emerging in the soft X-ray range, where longer wavelengths do not couple directly to interatomic distances but rather to magnetic and chemical structure at nanometer and above dimensions that is of fundamental importance in many areas as discussed in Section 2. Early examples in the soft X-ray include the measurement of diffuse scattering away from the specular reflection from a magnetic film [46,48]. These studies suggest that the magnetic and chemical roughness that produce this diffuse scattering are not necessarily identical. In another example, the magnetooptical sensitivity in the specular reflectivity intensity from a magnetic multilayer has been used to observe an



Orbital
Moment



Spin
Moment

Co thickness t (monolayers)

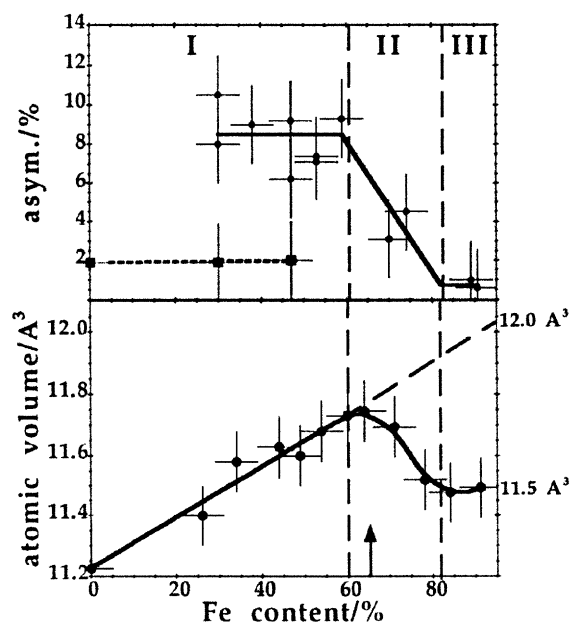


Fig. 7. Changing structure and magnetism with composition in six-monolayer thick $\text{Fe}_x\text{Ni}_{1-x}/\text{Cu}(1\ 0\ 0)$ films are revealed using photoemission in conjunction with magnetic linear dichroism. The composition dependence of the dichroism at the iron and nickel 3p levels (top) and of the atomic volume of the films (bottom) reveal a clear correlation between structure and magnetism. (from Ref. [171]).

antiferromagnetic multilayer interference peak unobservable with charge scattering alone [46]. Other examples emphasize the compatibility of scattering techniques with variable applied magnetic fields to sense switching of individual layers in layered structures, providing information on the degree of magnetic correlation between layers [177,178]. The above examples have relied on scattered-intensity measurements. However, measurements of polarization changes in reflected (scattered) and transmitted beams have also been demonstrated, thereby extending Kerr and Faraday rotation spectroscopies and hysteresis measurements into the

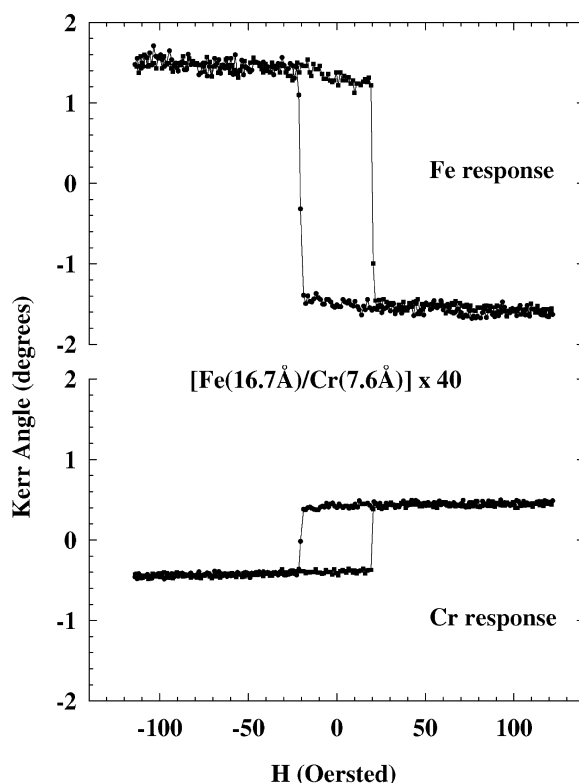


Fig. 8. Longitudinal X-ray magneto-optical Kerr rotation hysteresis loops reveal the individual magnetization response for iron and chromium layers in a polycrystalline Fe/Cr multilayer. In this sample the Fe layers couple ferromagnetically and the Cr layers possess a significant moment oriented opposite to that of Fe. The net Cr moment results from the interfaces where a significant number of Cr moments are uncompensated, possibly due to intermixing with Fe to yield ferrimagnetic local order. Obtained using a low 2° grazing-incidence angle, these curves are primarily sensitive to the in-plane magnetization in just the top Fe/Cr bilayer, while measurements at higher angles in reflection or in transmission would weight the curves with significantly increased contributions from deeper layers (from Ref. [49]).

VUV/soft X-ray range [49,50]. Fig. 8 shows hysteresis loops of iron and chromium in a polycrystalline iron/chromium multilayer, revealing that chromium possesses a net moment opposite to the iron

Fig. 6. Origin of the magnetocrystalline anisotropy illustrated by XMCD results for a Au/Co/Au wedge sample. The wedge shown has an in-plane easy axis at the thick end and an out-of-plane easy axis at its thin end. The measured angle-dependent orbital moment is found to become increasingly anisotropic towards the thin end where it strongly favors a perpendicular direction and redirects the spin moment from an in-plane orientation, the one favored by the dipolar coupling of the isotropic atomic spins (shape anisotropy), to the unusual perpendicular direction. The measured size of the independently determined isotropic spin moment is also shown (see Ref. [36]).

moment in these structures. As in both visible magnetooptics and hard X-ray magnetic scattering, the ability to measure polarization as well as intensity of scattered beams is often essential in rigorous interpretation of data.

Careful consideration of the magneto-optical properties and experimental details will allow these emerging scattering techniques to be applied with varying depth sensitivity. Simply increasing the grazing-incidence angle results in increased penetration depth, and so changes in hysteresis loops with angle would indicate changing magnetization of a given element with depth. A higher degree of depth sensitivity will be obtained by using optical standing waves to probe the magnetization across buried interfaces. Standing waves produced by the interference from multilayer structures readily modulate diffuse scattering [179], suggesting many extensions into magnetic multilayers near spectral resonances. Opportunities exist to apply these capabilities to study magnetism of the buried layers and interfaces that are often critical in defining magnetic properties of interest for applications. Examples include studying buried layers (including spacer layers) in spin valve and MTJ structures and the antiferromagnetic/ferromagnetic interface where the mechanisms of exchange biasing are thought to be associated with uncompensated interfacial spins of the antiferromagnetic layer [125]. It should be possible to study magnetic fluctuations and inhomogeneities associated with reversal processes and phase transitions with scattering, possibly in a time-resolved fashion.

4.3. *Spin-resolved electronic structure*

Following inverse photoemission studies [26,27], spin-polarized photoemission studies of noble-metal thin films deposited on ferromagnetic substrates [180,181] provided direct evidence that spin-polarized quantum-well states are available to mediate the coupling in the associated magnetic multilayers. These studies focused the discussion of the proposed mechanism of oscillatory coupling on the relative binding energies of the spin-dependent band gaps in the ferromagnetic layers, thereby providing a mechanism for oscillatory coupling in terms of spin-polarized quantum confinement of

electrons in the noble-metal films resulting from the spin-polarized band structure in the ferromagnetic parent materials. When the spacer-layer thickness is varied, the quantum-well states move up to the Fermi level, at which point the total energy of the system reaches a local maximum. The energy of the system is reduced by switching to antiferromagnetic alignment which moves the band gap away from the Fermi level and reduces the confinement of the quantum-well state.

These seminal early measurements have recently been extended at the ALS to include moderate spatial resolution and to investigate coupling between quantum-well states in different spacer layers [182]. These extensions are possible only with small focused beam spots (50–100 μm at 1 : 1 focusing), which result directly from the brightness of the ALS, in conjunction with in situ grown, wedged samples to provide an entire range of sample thickness for study from one deposition session. High flux and resolution ($> 10^{12}$ photons per second at a resolving power of 10 000) then enable combining high-resolution photoemission measurements with lateral scanning of the sample to obtain spatially resolved maps of photoemission spectra. In this case, the diameter of the focused beam spot and film-thickness gradient translated into a resolution of 0.5 monolayers in layer thickness. Single-wedged layers of Co/Cu(0 0 1) have been studied, as have more complicated structures consisting of double-wedged structures of Cu/Co/Ni/Co/Cu(1 0 0), where the copper and top cobalt layer were wedged to detect the interference of the quantum well states in the copper and nickel layers. The results shown in Fig. 9 clearly indicate how the intensity of the copper quantum-well state depends on the cobalt thickness (copper-nickel separation), thus identifying this interference effect. The results strongly support the quantum-well model of the oscillatory magnetic coupling in the newly discovered GMR magnetic multilayers. These techniques can be extended by including circularly polarized incident radiation and/or spin-polarized electron detection to more finely resolve the spin-polarized nature of the quantum-well states. They can also be extended to many other classes of magnetic systems and problems.

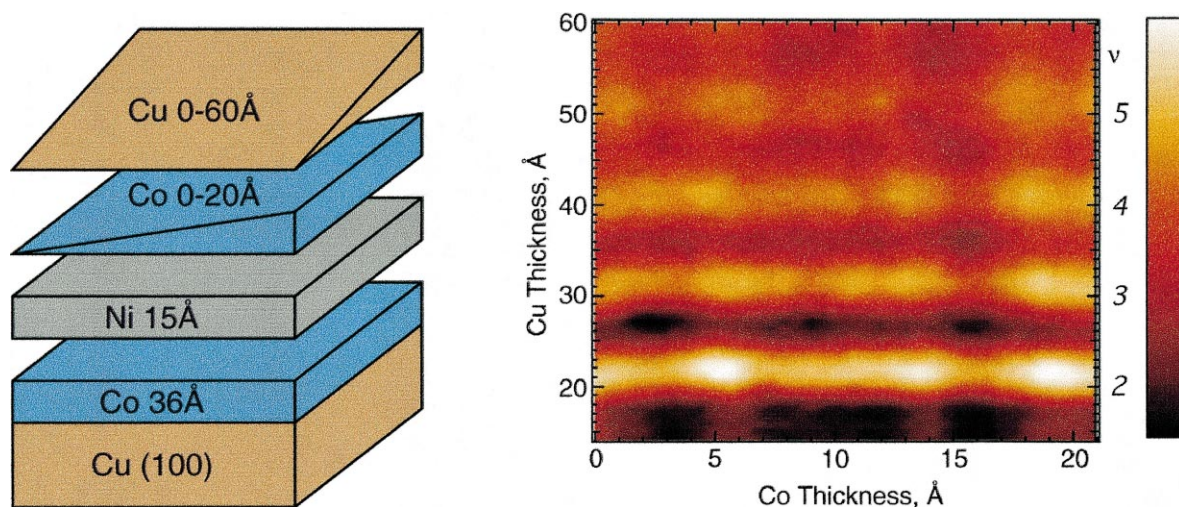


Fig. 9. Interference between quantum-well states in different layers of magnetic multilayers is shown in the image of photoemission-intensity modulations with varying thickness of two different layers (see sample schematic). In addition to the double-wedged sample (grown in situ), an intense X-ray spot 50–100 μm in size made possible by the undulator brightness was needed to observe these features. The clear dependence of the interference on multiple film thicknesses provides strong support for a model in which spin-polarized quantum-well states mediate oscillatory coupling in magnetic multilayers that in turn exhibit giant magnetoresistance (from Ref. [182]).

The colossal-magnetoresistance manganites and related oxide materials exhibit a range of interesting phenomena as a function of doping that demand a microscopic, and ultimately atomic, characterization of structure, electronic structure, and dynamics. At low temperature below the metal-insulator transition, they are predicted to exist as half-metallic ferromagnets. A recent spin-polarized photoemission study has provided the best experimental evidence that this is the case, as shown in Fig. 10. More detailed studies of these materials will require spin-polarized studies with energy resolution comparable to kT_C , where T_C is the Curie temperature. In the case of $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ for example, this resolution would be of the order of 25 meV, and it can be obtained at a third-generation source such as the ALS by combining high-resolution photoelectron spectrometers with micro-Mott or other spin detectors of improved-efficiency. Additional important information concerning these materials will come from companion core-level photoemission studies with variable photon polarization and spin resolution; for example, it should be possible to resolve the two types of manganese ions and to

study their short-range magnetic order separately, perhaps using spin-polarized photoelectron diffraction. Clearly, photoemission techniques will continue to evolve to higher spatial, energy, angular, spin, and time resolution so as to provide spin- and momentum-resolved electronic structure and magnetic structure for many important problems in magnetic and other materials.

In most cases photoemission and inverse photoemission are the techniques of choice for studies of electronic and spin structure, owing to their high energy and momentum resolution and, in the case of core levels, also their element specificity. In cases of buried layers and interfaces, X-ray emission spectroscopy (for a review see Ref. [183]), a technique that has already undergone a renaissance at the ALS, offers unique capabilities. Similar to valence photoemission, the X-ray emission technique probes the filled electronic states, but because of the initial core hole, it does so with elemental specificity. Thus, X-ray emission spectroscopy allows an investigation of chemical bonding and hybridization effects. Application of this technique to cobalt/copper magnetic multilayers [120], for example, has shown the hybridization of the cobalt

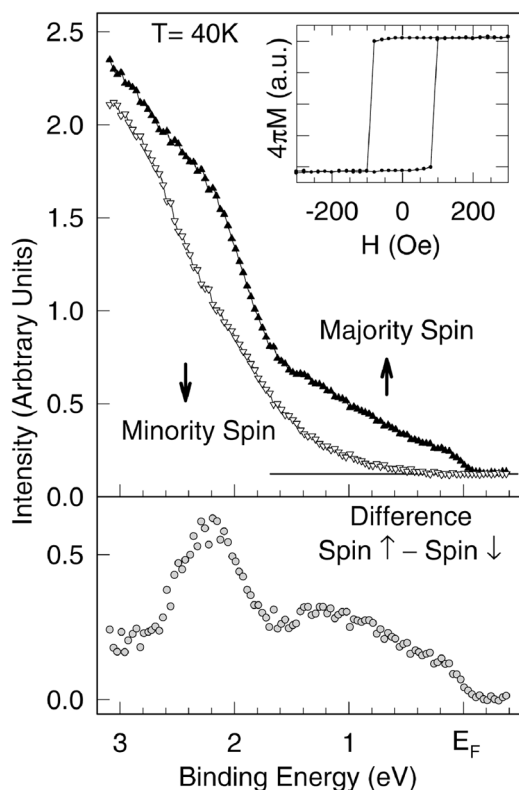


Fig. 10. Spin-polarized photoemission spectra of a 1900-Å thick film of $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ taken at $T = 40\text{ K}$ ($T_C = 350\text{ K}$). The photon energy and experimental resolution were 40 and 0.2 eV, respectively. A magnetic pulse coil with a magnetic field of about 200 Oe was used for magnetization of the sample. The inset shows the magnetization (M) versus applied magnetic field (H) hysteresis loop, which was obtained by monitoring manganese L_2 -edge absorption of circularly polarized incident light (from Ref. [28]).

and copper densities of states at the interfaces, giving direct evidence for the origin of the magnetic moment on copper interface atoms found by XMCD absorption spectroscopy.

In addition to the extensive capabilities of photoemission for characterizing the electronic structure of magnetic materials, XMCD and other core-level spectroscopies are having an impact on magnetism through probing electronic structure. Examples relating to anisotropy are given above. Other examples include alloys, oxides, and compounds in which XMCD spectra of different elements are probed. Induced moments on nominally non-

magnetic elements in multilayers and alloys have been observed by XMCD absorption. Moments have been observed on oxygen in CrO_2 [184] and the colossal-magnetoresistance compounds $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ [185]. These intriguing observations have direct relevance to the microscopic exchange mechanisms that are thought to mediate magnetic interactions. This atomic-scale understanding of magnetism in multicomponent structures may be used to test proposed exchange models, in the CMR materials for example, and to motivate further theoretical research in a wide range of materials. Another example is understanding the origin and nature of carrier-induced magnetism in compound magnetic semiconductors. Capabilities of probing both surface and bulk or buried-layer/interface properties are again important considerations in these types of studies.

While demonstrated potential and new opportunities clearly exist to study spin-resolved electronic structure with core-level XMCD spectroscopies, important questions remain about the core-level spectroscopies themselves. The detailed mechanisms of the spectroscopies are quite complicated and may influence the very properties of interest to investigate. In particular, the large MO signals near core levels result from processes in which a core hole is created, which may perturb the final states whose unperturbed spin and orbital polarization are of primary interest. The X-ray physics at ultrashort time scales associated with transition processes and the many-body responses to the creation of the core hole are thus critically important in assessing the degree to which the measurement distorts the system. This implies an essential close coupling of the theory of such experiments with the next generation of such experimental work.

It is also worthwhile to note that the development of new techniques for probing the atomic and electronic structure of magnetic materials should be possible at a facility like the ALS. One early example of this is the interatomic multiatom resonant-photoemission effect that has been observed for the first time in several metal oxides (MnO , Fe_2O_3 , and $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$) [186]. This effect, which couples the photoelectron excitation of one core level (e.g., oxygen 1s in MnO) with a resonant

absorption on a neighboring atom (e.g., manganese 2p in MnO), has the unique potential for identifying which atoms are neighbors to a given atom, for doing spectroscopy only on interface atoms that have some degree of hetero-atomic bonding between homogeneous layers, for studying magnetism via XMCD effects in the resonance, and for studying more deeply buried interfaces by using secondary X-ray emission processes for detection.

4.4. Time domain/dynamics

Some early experiments using synchrotron radiation [187] and accelerator-produced electron beams [79] have studied time-domain issues, but this is a largely unexplored area in the context of synchrotron radiation where interesting opportunities may exist. The issues associated with dynamics are of central importance to research in magnetism and magnetic materials as a whole, where pushing from nano- to picosecond time scales are especially important. One general limitation at these time scales is the ability to produce intense, picosecond magnetic-field pulses.

Nanosecond XMCD spectroscopy has been demonstrated at the European Synchrotron Radiation Facility, where 25 and 50 ns magnetic-field pulses with an amplitude of 0.1 T were applied repeatedly to GdCo₂ amorphous thin films. Data were collected over three minutes, averaging over 80 million minor hysteresis cycles. Time resolution in the XMCD measurement was achieved through synchronization and delay of electron bunches with respect to the field pulses. The magnetization oscillated in time after the field pulses were applied over the probed area of 30–100 nm², going first to negative values [187]. In a second experiment, the dynamics of magnetization reversal at surfaces has been studied using soft X-rays emitted from only one electron bunch. This novel time-resolved surface magnetometry measures the spin polarization of the total photoejected electron yield by means of Mott scattering. The first results obtained with this technique are concerned with the magnetization reversal of iron ultrathin films deposited as amorphous low-coercivity ferromagnetic ribbons [188].

4.5. Magnetic spectromicroscopy

The ultimate goal in magnetic microscopy would be the direct observation of atomic spins and their motions and the correlation of the orientation and size of the atomic spin and orbital magnetic moments with the atomic structure and composition of the sample. The accomplishment of this goal would revolutionize our fundamental understanding and lead to new designs of technological devices. In many ways, these goals of magnetic microscopy underlie many of the research frontiers summarized in Table 1. It is for this very reason that much effort has been exerted to push the magnetic resolution towards the atomic limit.

To put VUV/soft X-ray studies in proper perspective requires noting that magnetic microscopy can be carried out with several different and competitive approaches such as Lorentz microscopy, Kerr microscopy, secondary electron microscopy with polarization analysis (SEMPA), spin-polarized low-energy electron microscopy (SPLEEM), and magnetic force microscopy, to name a few [189]. To date the best methods offer magnetic resolutions around 10 nm. In this environment, the question arises as to whether synchrotron-radiation-based magnetic microscopy can make unique and important contributions.

It is already clear from the discussion in Section 3 that, in principle, magnetic microscopy based on core-level spectroscopies (spectromicroscopy/microspectroscopy techniques) offer several unique capabilities, most important of which are elemental and chemical- state specificity, the ability to separate the spin and orbital magnetic moments, depth sensitivity varying from surface to bulk, the ability to image antiferromagnetic domains (for first results see Ref. [190]), and the ability to image in the presence of large magnetic fields [191]. An example is shown in Fig. 11 where the field-dependent domain structure is observed at a lateral resolution of 30–40 nm [191]. Despite these various unique capabilities, X-ray-based spectromicroscopy has had little impact on research in magnetism and magnetic materials in part because its resolution has historically lagged behind the leading techniques. Also, ‘home based’ techniques are

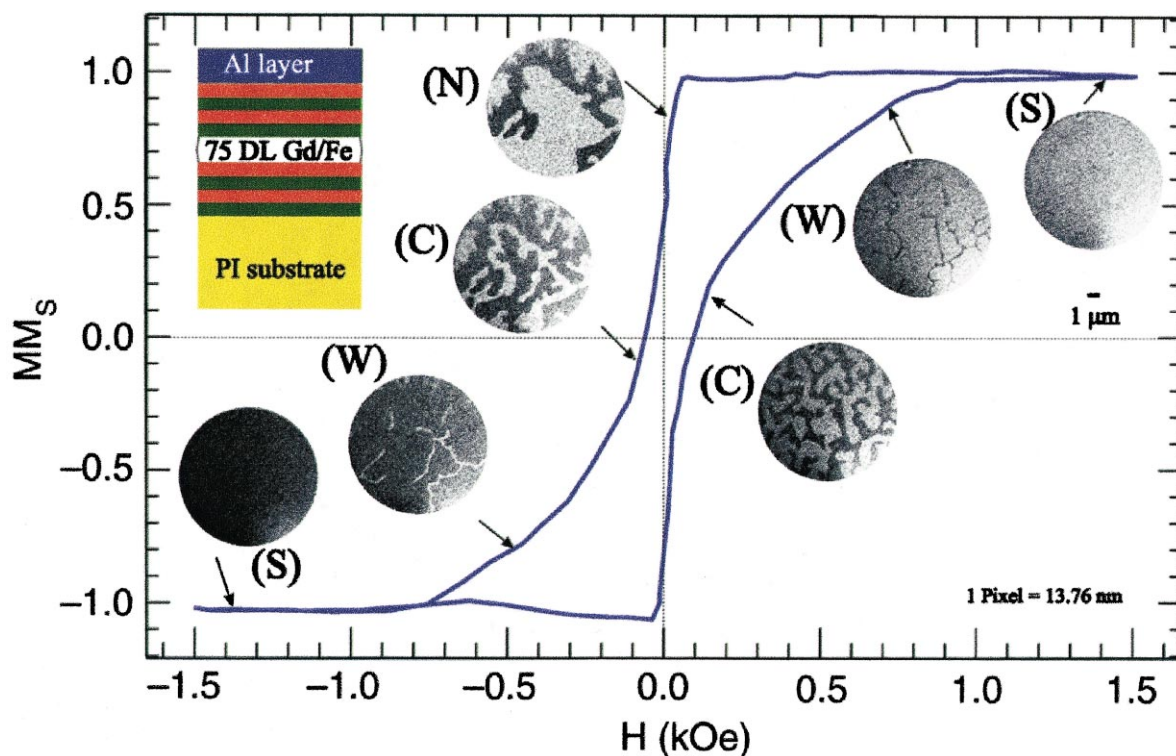


Fig. 11. Magnetic domain pattern of a Gd/Fe multilayer film measured with circularly polarized X-rays at the iron L_3 edge as a function of an applied magnetic field, using a transmission X-ray microscope. The domains have magnetization directions perpendicular to the film. The magnetic resolution is between 30 and 40 nm (from Ref. [191]).

more accessible and the instrumentation is less expensive.

Looking ahead, in order for magnetic spectro-microscopy to have an impact on magnetic materials research, unique capabilities of the X-ray techniques must be brought to bear on important problems. One technical improvement here is to push the lateral resolution below 10 nm. In fact, resolutions of about 2 nm appears possible with an aberration-compensated PEEM microscope [164]. Such a microscope, if available in a timely fashion, could have a significant impact on magnetic-materials research. For example, it would help in understanding the stability of the magnetic bits and the magnetic structure of the transition regions between the bits in magnetic-recording media, as shown in Fig. 12. Increases in storage density require the design of new materials with domains that are stable to smaller dimensions (i.e., by somehow

avoiding the superparamagnetic limit through materials engineering) and that have reduced transition widths (about 10 nm). Such materials will most likely consist of grains of reduced size (< 10 nm) that are separated from each other at the grain boundaries by a magnetically passive material. The ability to obtain element-specific magnetic images of such materials will be of great importance.

A second area of opportunity is to make use of the elemental sensitivity, penetrating power, and large contrast to directly image domains and magnetization correlations into the depth of multilayer structures. It appears that only X-ray microscopy techniques can directly image magnetization in different layers (including possibly buried interface layers) by tuning to core levels of elements distinct to each layer. So these techniques should have impact on characterizing three-dimensional

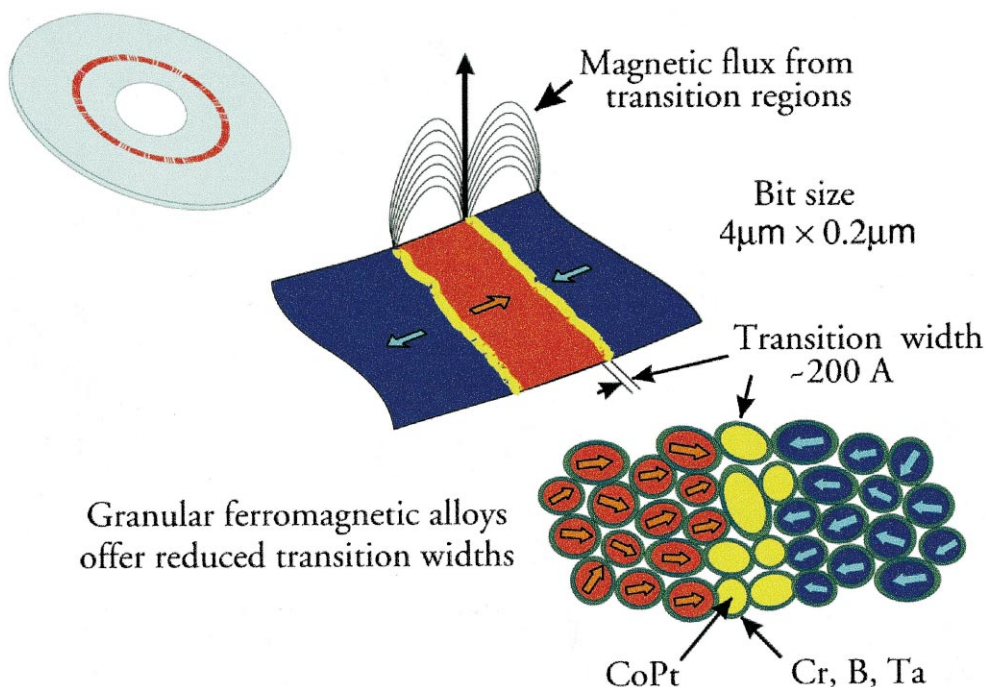


Fig. 12. Advanced computer disks consisting of granular magnetic materials like CoPtCr with admixtures of boron or tantalum in order to minimize the transition width between the magnetic domains. In the disk material, the grains are believed to be coated by a nonmagnetic shell that reduces the magnetic coupling between the grains. A small transition width is required in order to achieve a high magnetic-flux density in the direction perpendicular to the disk surface, as shown. The flux from the spinning disk is sensed by the spin-valve magnetic read head shown in Fig. 2 (figure courtesy of J. Stöhr, IBM Almaden Research Center).

magnetization distributions in nanostructures of both fundamental and applied interest. Many questions relating to the interaction of one magnetic layer with another can now be studied with spatial resolution of the magnetization in the individual layers. The transmission geometry facilitates quantitative magnetization imaging, which will be valuable in testing micromagnetic theories. Imaging in applied fields then enables observation of how the reversal process proceeds in different layers, or alternatively allows spatially resolved hysteresis loops of buried layers to be measured. Such capabilities may be important in the development of magnetic memory elements.

A third area of opportunity lies in the ability to image antiferromagnetic domains by means of linear-magnetic-dichroism microscopy and, by a change to circular X-ray polarization, ferromagnetic domains in the same sample. Such studies

may well hold the key to solving the important exchange-anisotropy problem discussed in Section 2.1.2. Here and in many other applications, the ability of X-ray dichroism microscopy to probe buried layers and interfaces is essential.

5. Conclusions and recommendations

The importance of magnetism and magnetic materials to society through technological and economic factors is considerable. The magnetic storage industry alone is about one-third as large as the semiconductor industry. In addition, the possible revolution in permanent-magnet materials with the advent of 'spring' magnets will have overwhelmingly positive repercussions throughout society. Evolutionary growth in materials-synthesis capabilities is currently linked with vigorous basic

research into wide-ranging areas of magnetism and magnetic materials, with clear potential to continue to impact science, technology, and society. The increasing complexity of these materials and the novel phenomena they exhibit require increasingly sophisticated experimental probes to gain fundamental understanding. Of particular importance is the reduction in size of the fundamental elements to the nanometer or even atomic scales.

It is clear that a third-generation VUV/soft X-ray synchrotron source such as the ALS offers new opportunities to positively influence the fields of magnetism, magnetic materials and their application in many ways. Both established and emerging VUV/soft X-ray synchrotron-radiation spectroscopic techniques couple directly and with large signals to the electronic and spin states containing the physics responsible for these unique properties. These techniques can directly probe the valence electrons that lead to magnetism, and through core electronic excitations they provide an atomic-scale sensitivity to magnetism. Together they provide, with spatial sensitivity at the sub-angstrom level (e.g., through photoelectron and X-ray diffraction methods) and at the nanometer level and above (e.g., through scattering and spectromicroscopy), clear opportunities to move beyond classical models of magnetism (e.g., those based upon the usual Landau–Lifshitz–Gilbert methods) and to establish an understanding of magnetism at a more microscopic and quantum-mechanical level. If time resolution down to the nano- and picosecond domains can also be incorporated into these techniques, it will dramatically enhance their ability to impact the understanding of magnetism at this fundamental level.

Of crucial importance for increased impact is an enhanced coupling of the ALS to the magnetism and magnetic-materials community. This is especially important precisely because magnetism and magnetic materials are very well-established fields, with many groups actively engaged in such research at their home institutions. On the other hand, the unique capabilities offered by third-generation sources of VUV/soft X-ray synchrotron radiation require dedicated development of instrumentation and techniques, as well as on-site sup-

port for outside users by experts who may be separated from mainstream magnetism research groups. This working group thus recommends the establishment of an ALS outreach program to bring these groups together in order to help ensure that the synchrotron-radiation resources are brought to bear on problems that will have a maximum impact in the true frontiers of research in magnetism and magnetic materials. Such interactions are encouraged with the magnetic-storage industry, the permanent-magnet industry, other industries making use of magnetic materials, and university and government laboratory groups.

The ALS should work closely with user groups to push the promising magnetic-spectromicroscopy efforts towards their ultimate nanometer-scale resolutions and to develop other unique spectroscopic, diffraction, and magnetooptic capabilities made possible by the brightness of the ALS. Because of the ALS brightness, spectromicroscopy is a unique strength of the facility, and every effort should be made to exploit this capability. Maximum impact in this area will require the establishment of a user base with deep roots in the magnetics community. The ALS is encouraged to form and work with such a user group to raise the funds necessary for the required instrumental advances towards the ultimate lateral-resolution limits. It is also clear that state-of-the-art facilities for various spectroscopic techniques involving both photon-in/electron-out (e.g., valence and core photoemission with spin resolution and variable polarization) and photon-in/photon-out (e.g., X-ray absorption, scattering, and emission) are necessary complements to maximize the unique impacts of the ALS in magnetism.

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